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## Composite Material Impregnation Unit

S. P. Wilkinson  
J. M. Marchello  
N. J. Johnston

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# Composite Material Impregnation Unit

## 1.0 INTRODUCTION

As part of the Polymer Composites Materials Research Program, a multi-purpose prepregging machine has been designed and built for NASA Langley Research Center. The machine is capable of impregnating continuous fibers with high performance polymeric resins being developed at NASA Langley and has a number of advantages over existing units due to its various modular components. Each of the modules can be used individually or simultaneously depending on the required prepregging method.

A reverse roll coater provides the ability to prepare thin films from typical hot melt thermoset formulations. Also, if necessary, the design allows direct fiber impregnation within the reverse roll coater gaps. Included in the impregnation module is a solution dip tank allowing the fabrication of thermoplastic prepregs from solution. The proceeding modules within the unit consist of four nip stations, two hot-plates, a hot-sled option and a high temperature oven. This report describes the benefits of such a modular construction, discusses the various processing combinations available to the prepregger, and presents operation and safety information.

An analysis of the design and operating features of the prepreg tape machine was conducted to provide insight into the full utilization of the machine. In addition to a description of individual component parts, governing equations for material conservation and heat and mass transfer were developed to describe steps in the prepreg process. These relationships will be utilized in standard engineering operations to define the operating protocol for the machine. The primary goal of this analysis was to provide the basis for directing future efforts into understanding the science and engineering of the production of high quality prepreg.

The following list presents the various aspects of prepreg quality and operating specifications that need to be addressed when performing an engineering analysis on the tape machine. Variables presented in this list may be split into two types, those that affect the product specifications required by the vendors, and those internal design/operating variables which govern the manufacture and production costs of the machine.

## **Material Requirements and Machine Operating Variables**

### **Prepreg Specifications**

Resin content  
Resin distribution  
Fiber Areal weight  
Splits  
Tack  
Drape  
Solvent content  
Voids

### **Internal design/Operating Variables**

#### **Machine Operating Variables**

Running speed  
Oven Temperatures  
Oven air flow rates  
Metering rod gaps  
Nip roller gaps  
Nip roller temperatures  
Nip roller pressures

#### **Material Properties**

#### **Solution properties**

Polymer properties  
Solvent properties

## 2.0 PREPREGGER OPERATIONS MATRIX

To produce a prepreg material that possesses the desired specifications, a knowledge of the machine design and operating procedures which relate to meeting these specifications is required. The following matrix correlates the prepreg specifications with the various operating units that form the prepreg tape machine. The top five rows are functions of the applied nip pressure and temperature. The bottom three rows are functions of temperature only.

PREPREGGER MODULAR COMPONENTS									
	Creel* and comb*	Fiber Tension	Film or Dip tank	Nip #1	Hot plate	Sled option	Nip #'s 2 and 3	Oven	Nip #4
Resin content			√	√		√	√		*
Resin Distri- bution		√	√	√		√	√		*
Fiber Areal Weight	√			√		√	√		*
Splits		√	√	√	√	√	√	√*	√*
Voids		√	*	√	*	√	√	√	*
Fiber damage	√	√		*		*	*		
Tack					√	√		√	
Drape					√	√		√	
Solvent content					√	√		√	

### Key

√ = Primary factor - Such modules are directly responsible for affecting the given prepreg specifications



\* = Secondary factor - These factors show how the individual modules may indirectly affect the prepreg specifications.

The process variables utilized in the design and operation of the tape machine are discussed in the following sections. Modular components of the prepregger are illustrated in Figure 1 and are discussed in subsequent sections.

### **3.0 PREPREGGING METHODS AND ANALYSIS**

In this section the technical interpretation of the prepregging operations of which the NASA machine is capable are described and basic calculations related to these operations are presented. In some cases scientific interpretation of the phenomena involved in prepregging are also introduced and conceptually described as reference points for further research and development. Computations presented here are those needed for basic machine operation and make use of the following variable definitions.

#### **3.1 Variable Definitions**

FAW = Dry fiber areal weight

RAW = Resin areal weight

PAW = Dry prepreg areal weight

(PAW)<sub>wet</sub> = Prepreg areal weight with volatiles

W<sub>f</sub> = Fiber areal weight fraction of PAW or (PAW)<sub>wet</sub>

W<sub>m</sub> = Matrix weight fraction of PAW or (PAW)<sub>wet</sub>

W<sub>solvent</sub> = Solvent weight fraction of (PAW)<sub>wet</sub>

W<sub>solid</sub> = Solid weight fraction of solution (i.e. 40% solid solution = 0.40)

t = Dry prepreg thickness

(t)<sub>wet</sub> = Prepreg thickness with volatiles

ρ<sub>c</sub> = Prepreg density

(ρ<sub>c</sub>)<sub>wet</sub> = Prepreg density with volatiles

Ends = # of fiber spools

y = Fiber yield

w = Prepreg width

## 3.2 Reverse Roll Coating

### 3.2.1 Film Casting

The off-line film coating process is most commonly employed to form a film using hot-melt thermosets. The unit uses three rollers as illustrated in Figure 2, with resin being applied to the preset gap between the backing and metering rolls. Resin is carried through the metering gap and pressed against the release paper surface which travels on the third roller in a counter direction. A thin resin film adheres to the paper surface after the resin profile has been sheared due to the opposing motion of paper and resin. This shear splitting effect must be taken into account in calculations for the metering gap dimensions. Unfortunately, little information exists on the shear thinning behavior of resins. As numerous materials, which vary in their shear thinning nature, may be utilized in the film formation process, research on materials' shear behavior is an area that would aid in the further development of reverse roll coating technology. The prepreg's resin content is primarily controlled by the film's resin areal weight as indicated in Appendix Ib.

The pre-cast films are stored in a refrigerator until used for prepregging fibers in a separate process as illustrated in Figure 3. Quality prepreg is often made using the pre-cast film processes when using conventional thermosetting addition type polymers. However, the more novel resins being prepared present a wide variety of flow and adhesion properties. In these cases parameters such as film quality, film storage, and solvent content within the film (required for film processing) become important issues. Often the film's adhesion and release properties from backing paper are a concern in producing good quality prepreg. Certain polymeric materials, such as 30-40% w/w solutions of polyamide acids, may form islands or pools of resin on the surface of the release material when heated or when being nipped with the dry carbon fiber web. This would produce an unfavourable non-uniform resin distribution across the prepreg width. The adhesion/release material behavior of new resin systems therefore requires further investigation.

### 3.2.2 Direct Impregnation at the Reverse Roll Coater

An alternative to the off-line paper coating process, described above, is that of on-line coating, which is somewhat similar to hot-melt prepregging. In this instance the fiber is impregnated "on-line" within the gap between metering roll and applicator roll, such a process is illustrated in Figure 4. The "on-line" process is typically performed with resin solutions that possess high viscosities, and can not be impregnated using the solution dip tank process. Top and bottom release papers are

brought into the metering gap along with the fiber web and resin system. Impregnation takes place at this junction with the pool of resin contained in the nip gap by dams at each end. This process may be suitable for impregnating high temperature performance oligomers that melt with viscosities in the order of 30,000 to 100,000 cp. Using a typical lubrication analysis, it can be shown that very high pressures can be attained for the impregnation of such material systems, providing the gap at the reverse roll coater is kept to very small dimensions (1). This method of impregnation deserves further attention as new oligomeric systems are continually being developed.

### 3.3 Solution Dip-Tank

Solution prepregging, like hot-melt, involves passing the fiber web through a liquid resin. In solution prepregging, the resin tank contains a liquid which contains a polymeric material dissolved in a suitable solvent.

The solution dip tank and resin level metering control bars are shown in Figure 5. The gap between the metering rods is adjusted to control the amount of resin added to the fiber web. The material balance equations, Appendix 1c, describe the general procedures used to estimate the required gap setting.

The gap setting at the metering rods provides preliminary control over the resin solids content. The concentration of polymer in solution will affect the viscosity (as will the polymer molecular weight), these parameters alter the polymer solution characteristics and change liquid behavior in the dip tank. Since the effects of viscosity and density of the solutions on tow bundle impregnation are not known, the actual gap required may be larger. This is another area where research would be useful in improving the design and operation of the machine.

For some solutions, such as polyamide acids or polymers from monomeric reactants (PMR type resins) the above calculations also need to take into account extra volatiles that are released from these resins when they are further processed to form their imide moieties. These volatiles are not residual solvent and must not be treated as such in calculations. Polymers that may change in their final weight in such a manner need to be studied with respect to just how much mass they will lose. When this has been done, the desired matrix weight fraction ( $W_m$ ) can be adjusted in the calculations. For example, if LaRC™RP46 (a PMR type resin system) was to be prepregged and its desired final weight percent solids was to be 33%, then an additional 6% should be added to make 39% the  $W_m$  value used in the metering bar calculations. This will take into account the methanol that is released when the BTDE ester is cyclized to form the dianhydride, and the subsequent water which is released

when this dianhydride reacts with the diamine to form the amide acid and then cyclizes to form the imide. The 6% is calculated by estimating that the LaRC™RP46 resin will be reduced in weight by 17% and that the prepreg product will be 2/3 fiber and 1/3 resin, (hence 1/3 of 17% is ~6%). Appendix Id presents an example that shows how the 17% extra volatiles is calculated, knowing the chemistry of the LaRC™RP46 resin components and the percent solids concentration of the solution to be prepregged.

Viscosity control at a particular polymer concentration is an important operating variable. Too high a viscosity can create a large shear drag on the prepreg web, which in turn may create fiber damage. Temperature control has been installed on the dip tank, and, providing no unusual solution properties are found upon heating, viscosity can be controlled through heating the solution. One problem that may occur with the dip tank is skin formation on the liquid surface due to the effects of humidity which cause precipitation of the polymer from solution.

A dry fiber web entering the dip tank allows air to enter the solution and creates foam formation. The foaming phenomena may be a limiting part of the prepregging process and should be addressed in the future design analysis. The entrained air is squeezed out at the metering rods, but a backwash effect of flow back into the tank suggests that this may be the limiting step when determining the optimum tape speeds. Entrained air bubbles raise the liquid level in the tank. The net amount of air present in the tank solution as small bubbles depends upon the difference between the rate with which the bubbles enter from the dry fiber web and the rate at which bubbles leave by rising to the surface, or, by being carried out with the wet tow web solution to the metering rods, where they are expelled from the liquid wave at the front of the bars.

During operation back flow of excess resin solution from the metering bars is exhibited as two standing waves in front of the bars, one above and one below the web. Liquid solution and air bubbles accumulate in these waves, which spread along the bar gap width to a point just beyond the edge of the web, where a portion of the solution passes through the gap and serves to join the two waves together. The resin solution wave level stabilizes by back flow of solution down the wet web and into the tank. This counterflow of solution results in a certain amount of accumulation of solution on the moving web, especially the top. In this way a dynamic equilibrium is established for the standing liquid waves, and accumulated solution, whereby excess resin carried up to the metering bars is matched by the return flow drainage. Resin back flow and air bubble elimination may be governed by tank and bar design. This is an area for further study.

The web speed also affects certain other variables including shearing forces in the transverse direction which can give rise to web splitting and resin squeeze out. Gaps, or splits in the web, are primarily dependant on surface tension and viscosity. They appear when web speed and transverse force overcome the wetting and flow properties of the solution. The fiber strengths and the effect of these shearing forces on fiber damage and web quality need to be addressed.

### 3.4 Resin Flow into the Fiber Web

The most critical process during prepregging is the impregnation of the dry fiber web by the resin. Depending on how the impregnation takes place, ie. through filming techniques or via the solution impregnation method, the lateral resin flow into the fibers results from the mechanical pressure being applied and the capillary rise pressure found within the web created by the fibers' packing geometry. A variety of resin flow models exist that may be utilized in conjunction with the analysis of the prepreg operation.

Darcy's law for flow in porous media is used to describe the impregnation rate in the thickness, Y, direction. The flow velocity, V, is a function of the resin viscosity,  $\mu$ , fiber bed permeability  $k_f$ , and the applied pressure, P.

$$V = \frac{k_f}{\mu(T)} * \frac{dP}{dY}$$

This equation for resin flow through the web is the starting point for most prepregging analysis. Additional relationships are needed to describe both the permeability and the resistance to impregnation due to the resin's viscosity.

Seferis (2,3) has discussed the fiber impregnation by introducing the concept of a prepreg flow number, PFN. This dimensionless parameter describes the interrelationship between temperature, pressure and production rate. It is dependant on the operating conditions of the prepregger and the geometry of a fibrous preform.

$$PFN = \frac{K P_{eff}}{\mu V Y_f}$$

K = Permeability

$P_{eff}$  = Effective pressure

$\mu$  = Viscosity

$V$  = Production rate

$Y_f$  = Thickness of a collimated fiber tow

In developing the PFN concept, Seferis (2) utilized Darcy's law and integrated the equation with respect to the fiber web thickness and rearranged the terms to obtain a ratio describing (in the numerator) resin impregnation by permeation, and, in the denominator, the viscous resistance to the resin flow. In this way the PFN number offers an indication of the quality of the resulting prepregs. A PFN greater than one implies that resin is easily impregnated into the fiber web and a ratio less than one indicates that the resistance to viscous flow is high and the prepreg is poorly wet out. An excessively high PFN number may be detrimental to the prepregging process, too much flow may create resin starved prepregs due to excessive squeeze flow of the resin.

The PFN concept analyses impregnation of fiber webs with hot-melt thermosets. In the case where impregnation occurs through a solution dip tank, additional parameters may need to be considered. Such parameters may include the change in the viscoelastic nature of the impregnated web as solvent is removed during the drying process.

In a solvent impregnated web, rich in solvent as well as polymer and fiber, the web becomes more elastic as solvent is removed during the drying process. The reduction in its viscous behavior and an increase in its elastic behavior provides a prepreg web that is more difficult to process. [A major problem occurs when the increase in the web's elasticity causes web splitting and creates gaps during the drying oven process, as will be discussed later. This is an area for future research.]

### **3.5 Uni-tape from Powder Coated Towpreg**

The fourth process that can be utilized to produce uni-directional carbon fiber prepreg involves the consolidation of powder coated towpreg. The towpreg material can be prepared off-line or on-line depending upon the design of the prepregger. The hot plates have the capability of being heated to 800°F. This introduces a substantially higher residence time for the polymer at such temperatures when compared with heated nip rollers that are limited to 425°F. Four rollers located on the hot sled in hot plate #2 (section 4.10) provide pressure. The pressures attainable at these contact points can exceed 1000 psi. In operation the sled travels back and forth over the web at speeds greater than the web speed.

The hot sled action tends to iron the towpreg material, and, with the correct combination of temperature, pressure and resin melt viscosity, a uni-tape product can be produced. An important process variable that needs further study is the type of release material that sandwiches the web and is used to protect the towpreg uni-tape during the hot sled operation. Aluminum foil or stainless steel sheet, approximately 0.001"-0.003", may be used in conjunction with the hot sled provided that the surface has a good release from the prepreg product. The release material thickness is also important in determining the pressures that can be reached to improve the consolidation of the uni-tape. Thicker material allows for higher pressures and helps to bring the product form to its net dimensions with a high degree of consolidation. Teflon® coated glass cloth may also be used as a release material, however, the Teflon® may be transferred to the prepreg surface during the process and could be a problem due to a contamination of the prepreg plies. The fiber tow tensions are another issue. They need to be high so that the tows do not buckle, wrinkle, or pucker during the process. If the tensions are too high, it will be difficult to spread the tows and achieve tow-to-tow adhesion. Further studies are needed to develop a processing window for the tow tensions. Section 8.0 describes the results obtained to date with prepreg manufactured via this method.

## 4.0 Prepregger Modules

The prepregger consists of various modules and is shown schematically in Figure 1. Each of these components is described in the following section.

### 4.1 Fiber Spool and Creel

Loading the fiber creel is the first step in prepregging. Spools of fiber tows are placed on the creel, each tow is threaded into the machine. Using the particular fiber of choice and measuring its fiber yield (Y), one calculates the number of ends necessary for providing a product with a specified fiber areal weight (FAW) and width (W). The following equation is used.

$$\# \text{ Ends} = \frac{\text{FAW} \times \text{W}}{\text{Y}}$$

Knowing the number of ends per desired width of prepreg, one calculates the number of tows per inch needed to obtain the desired fiber areal weight. Following

this calculation, a comb is selected that possesses a known tow count per inch due to the size of its gaps. The comb angle is calculated from the cosine of the ratio between the comb ends per inch and the desired ends per inch.

Loading the creel involves establishing a correct operational method for tow travel to ensure that the fibers do not cross over and bridge each other, (thus, avoiding unnecessary fiber damage during the prepregging operation). The following describes the procedure for the correct threading methodology.

The creel is split into two sides, each side consists of five rows of ten fiber spools. Located in front of the fiber creels are a series of horizontal bars. Fibers are passed over each bar designated for each row. That is, if three rows are used then fibers are passed over three corresponding bars. Likewise, all five bars are to be used, if all five fiber rows are to be threaded. The horizontal bars have vertical columns running down their length. Each fiber tow must exist within its own individual column. The threading of a particular fiber row over its designated bar must be performed by a certain method to ensure fibers do not cross when prepregging. The front tow of a row is located over its bar in the center column. As individual tows are threaded (from the front of the creel to the back), then each tow is alternated into columns at each side of the previously threaded tow. That is, each bar is threaded in columns from the center moving in a growing yet alternating fashion outwards.

Once each tow from its corresponding row has been threaded over each bar on either side of the creel, the comb at the impregnation module must now be threaded. Again, this is performed by splitting the threading procedure into two halves. Each half is threaded in the exact same way. The first tow is taken to be the most inner tow in the most inner column from the top threaded bar. This first tow is threaded at the center tooth of the comb. The second tow is now taken from the same column as the first, but now from the second bar down. It is threaded in the comb next to the first tow moving outwards. Likewise, the third tow is taken from the first most inner column but from the third bar down. The threading procedure is continued in this fashion until all the fiber tows from the first column are threaded. The threading operation now proceeds to the second column and tows are threaded in the same fashion, taking the tow from the top bar and moving downwards as each tow is threaded into the comb. This process is repeated until tows from all columns have been threaded into the comb from either side of the creel. The comb top is covered with a cap to prevent tows from becoming unthreaded and the comb is now set to the desired angle for prepregging.



Rolls of release material, usually paper, is mounted so that the top and bottom of the web is protected during various steps in the process. A number of release material applications and removal locations are available to the operator. The material is mounted on tension adjustable cores, tension is controlled by a pneumatic brake and tachometer

## **4.2 Impregnation Modules**

### **4.2.1 REVERSE ROLL COATER**

Typically this module is used to prepare precast films on release paper from hot-melt resin systems. The resulting films are brought into contact with the dry fiber web at the first nip roller station. Here, the resin is released from the paper and impregnated into the fiber web. As described earlier, the filming process is controlled by the gap dimensions at the coater where resin is sheared onto the paper surfaces as the paper passes between the applicator and metering roll, Figure 2. The rollers may be heated to control resin viscosity. Also, if desired, the dry fiber web can be passed through the roller gap and on-line impregnation can be performed at this position.

### **4.2.2 SOLUTION DIP TANK**

There are two dip tanks available for the solution impregnation operation. One tank is 15 inches in length and is required for prepreg tape 12 inches wide. The second is 5 inches in width and was specifically designed for prepreg tape 3 inches wide prepared from research-scale quantities. The minimum quantity of material for this tank is 320 ml and the maximum quantity is 900ml.

The dip tank assembly consists of three subassemblies. The first is the pan carriage and tow guides. The dip pan raises up to the fiber path, which is already threaded for impregnation.

The second subassembly consists of the dip pans and impregnation bars. The impregnation bars hang from two steel rods over which the dry fiber web is passed. As the impregnation bars are split in two, the web can be split in two and passed over two tiers, or, kept as one unit and passed over the bottom tier. A heating unit is located underneath the dip tanks which provides the operator with a viscosity control. By raising the solution temperature the viscosity may be lowered while maintaining a constant weight percent loading of solids in the solution.

The third subassembly is the metering rod assembly. The gap between the metering rods controls resin content and is the most important part of the solution

coating process. Resin content may be controlled by adjusting both the metering bar gap and/or the nip roller pressure. The gap is adjusted using wedge blocks that move a set of V-grooves. The adjustment screw is turned clockwise to adjust the metering gap, and feeler gages are used for monitoring the gap dimensions.

Further development studies are required in the following areas:

Firstly, the relationship between fiber impregnation and resin viscosity. Methods exist, as described earlier, for estimating the required metering bar gap to yield a prepreg product of a desired resin solids content from a solution of a known concentration. However, these calculations are based entirely on the respective fiber and solution densities and does not take into account the effect on tow bundle impregnation and resin pick-up from solution viscosity.

Secondly, the angle between the impregnated web and the metering bars. The amount of resin wipe-off, and resin backwash into the dip pan can have a significant effect on the resin pick-up into the fiber web. Solutions with low viscosities require larger than calculated gap dimensions to form desired solid contents on the prepreg. Further studies are required to estimate the importance on the angle between the web and the metering bars in determining the amount of wipe-off and backwash.

Thirdly, the effect of entrained air within the solution dip tank. Dry fibers entering the solution carry air with them. During steady operation these air bubbles continually enter and leave the solution. The population of air bubbles results in an elevated liquid level in the tank. The effect of this air bubble formation on the metering bar performance and fiber wet-out requires further study.

### **4.3 Nip Station #1**

The first nip station is used for the resin film transfer process for impregnating hot-melt materials that have already been pre-filmed (Figure 3). It is not used in solution coating because the solution wetted web is not amenable to nipping until initial drying has taken place in the hot plates and resin viscosity has been lowered. The rollers at the first nip station may be heated if desired. This gives control over the resin viscosity of hot-melt systems, an important parameter when determining the impregnation characteristics of an individual system.

All nip rollers in the machine are designed to provide gap dimensions that remain constant along the roller length. A tuning bar is located in front of each roller so that the wedge block at either side of the roller may be individually moved while maintaining the other stationary. Moving a particular wedge forward or backwards provides the operator with the ability to raise and lower each side of the roller. Feeler

gages are used to monitor the consistency of the gap dimensions. Two locks provide access to each wedge block movement and are situated on this bar, they can be individually released using the appropriate tools.

#### **4.4 Hot Plates**

At the beginning of a run, ovens and hot-plates are brought to operating temperature prior to starting the machine (and, thus; causing web movement). During operation heat from the hot-plates is taken away by solvent evaporation and convection to air, and by heating the web and paper as they pass over the plate. Since the prepreg web is very thin, its capacity to carry heat is small relative to the heat required for solvent evaporation. For solution prepregging, solvent is first removed at the hot plates.

During the initial devolatilization process, the bottom paper remains on the underside of the prepreg to protect the plates from resin build-up. Also, the paper may serve as a secondary solvent removal process by wicking away solvent and leaving polymer molecules behind. Solvent may wick to the paper edges and from there evaporate within the chamber. The following general analysis of devolatilization also applies to the oven chamber described later.

Heat transfer from the hot plate to the paper and prepreg, diffusion of solvent from inside the web to the surface, and devolatilization of solvent from the prepreg surface are all processes that need to be considered in describing the hot plate performance. Initially the web surface is wet with solvent. The rate controlling step with wet surfaces is evaporation of solvent into the air characterized by the "constant rate period" of drying (4). As solvent continues to evaporate the liquid level moves below the web surface and solvent vapor must diffuse from the web into the air. This is the "falling rate period" in drying analysis, (Appendix II).

The processes of heat transfer and solvent removal described in Appendix II, needs to be quantified and correlated in terms of machine performance and prepreg quality. This is an area of future study as web splitting is caused primarily by the drying process.

#### **4.5 Nip Station #2 and #3**

Achieving a desired fiber areal weight, while maintaining fiber uniformity across the width (requiring tow-tow adhesion and no splits or gaps within the web), is

an important aspect of product quality. The method for establishing a uniform gap was discussed in section 4.3 for nip station #1.

The nip stations #2 and #3 are essential in the devolatilization process and in eliminating splits and gaps formed in the prepreg web upon heating. At this primary control point, the following parameters that may be adjusted to eliminate splits and gaps are

1. Gap width between rollers, creating resin squeeze-out and lateral movement of the fibers
2. Back pressure to the nip rollers
3. Roller temperature (affects the flow properties of the prepreg)

Parameters that are controlled/adjusted in advance to the nip roller and contribute to the elimination of gaps and splits are

1. Web speed. This effects the pressure exerted on the web.
2. Solution concentration. A function of the hot plate temperatures and solution concentration in the dip tank. The polymer concentration changes the web elasticity.
3. Solvent content. Controlled by prior heating history.

The above parameters illustrate the need for developing a "gap elimination operating window" for solution prepregging. A basic understanding of the parameters that provide information about gap creation, and the material properties that are required for gap elimination to be successful during prepregging, are important criteria that must be established.

Temperature differences between the web and roller may create a temperature gradient throughout the prepreg from the internal regions to the surface. Consideration may be directed towards the use of release paper at these nip stations, and to how the temperature gradient is affected by such material. Different combinations may be imposed at the nip rollers, i.e. warm web and hot rollers, or a hot web and cool rollers. The main task of the rollers is to eliminate gaps. Although some further fiber wetting may occur, the majority of the fiber wetting process is believed to take place at the dip tank.

The process that occurs at these nip stations is a calendering process, (Appendix III). Utilizing equations based on the calendering process, the pressure

applied to the wet fiber web as it passes between nip rollers may be calculated from flow models.

One phenomenon which may occur at the nip stations, and most probably occurs within the ovens and hot-plates, is the "soaking up" effect of solvent into the porous paper, i.e. a chromatographic effect, leaving behind the polymer residing in the prepreg web. Hence, the paper may provide another method for solvent removal. This process would no doubt be a function of temperature and pressure. The phenomenon of "blotting" dry the prerpeg tape may present an entirely new approach to an environmentally sound, solvent removal process and deserves further theoretical and experimental investigation.

#### **4.6 Oven**

A critical unit within the prepregger is the oven because it provides control over the solvent content in the final product. The solvent level within the prepreg web needs to be monitored both at the entrance and exits. Variables that can change are oven temperature and air flow and web speed. Hence, residence time and temperature become important parameters to substitute into appropriate equations for heat and mass transfer that describe the solvent vapor removal, (Section 4.4).

The rate of air flow is a significant variable and affects the solvent removal process. Basically, hot air is driven into the chamber and heats the unit. Air flow opposes the direction of travel of the prepreg web. This has the effect of both heating the solution within the web to generate sufficient energy to vaporize the solvent, and also sweep away solvent molecules from the prepreg surface. The air flow and solvent evaporation can create a cooling effect in certain instances and should be a concern during the analysis. The mass transfer process of devolatilization consists of solvent diffusion through the prepreg thickness to the surface followed by evaporation.

Heat transfer is a primary issue in the oven's design and operation. Thermal energy is delivered to the oven by hot-air. During the prepregger's operation, air continues to flow to maintain the operating level, the direction being counter to the web movement. The two solid surfaces, the web and the oven walls can interchange radiant heat, i.e. act as black bodies. Due to air being transparent to infra-red radiation, the radiant heat is exchanged directly between the solid oven walls and the prepreg web. The oven space is heated by hot air flowing into the chamber and at the operation start up will bring the oven to the desired temperature. As shown in Figure 10, as hot air passes through the oven, heat is transferred by convection to the web and to the walls, therefore, the web receives heat from both hot air and the oven walls.

As in the case of the hot plates, an analysis of the heat and mass transfer in the oven must be conducted and related to web splitting and devolatilization.

#### **4.7 Nip Station #4**

This position provides the last opportunity to nip the prepreg web and to remove the remaining gaps and splits within the prepreg web. The roller gap width and the roller pressure exerted on the web are the major parameters that can be changed to attain such goals (section 4.5). The solvent content within the prepreg must be monitored and controlled prior to entering this nip. Hence, resin content, distribution, and fiber areal weight may be affected depending on the resin's prior history through the other modules that form the prepregger.

#### **4.8 Chill Plate**

The chill plate cools the web prior to take-up and storage. The chill plate is constructed from anodized aluminum and has parallel cooling channels to achieve an even temperature distribution. The chill plate is capable of temperatures below 60°F and is located between the nip and pull-roll stations. The chill plate is located 3/8" above the web line so that the web is in intimate contact with the plate. A one-ton water chiller with temperature control is supplied to maintain a steady plate temperature. The chill plate temperature is not monitored in the process.

#### **4.9 Take-Up Spool and Slitters**

The chill plate tends to "freeze" the resin and reduce flow. Therefore, any effects of resin impregnation at the drive motors are believed to be minimal. In turn, the lack of flow which the freezing imparts on the prepreg web does prevent resin loss through excessive squeeze-out and adhesion to the take-up paper. The chill plate may, therefore, have some affect on the final product quality in terms of reducing damage on wrapping when storing and taking up the prepreg.

#### **4.10 Hot-Sled Attachment**

The hot-sled module is designed to provide pressure needed to achieve impregnation during processing of high viscosity systems. The sled is capable of operating at temperatures up to 800°F. A schematic representation is depicted in Figure 11. The resin flow and heat transfer analysis will be similar to that presented earlier in Sections 4.4 and 4.5.

The hot-sled function is several fold.

1. Provides through the thickness penetration and wetting
2. Provides sufficient pressure to the fibers to create lateral movement and fiber nesting.
3. Causes web widening
4. Eliminates splits and gaps
5. Used when the nip stations #2 and #3 fail.

## **5.0 SAFETY FEATURES**

### **5.1 Emergency Stop Condition**

When hazardous conditions arise the red emergency stop buttons, located at several positions along the machine should be pressed to stop the machine. The machines pneumatic circuitry is tied into the electrical circuit that can be activated by pushing an emergency stop, or, when conditions occur, such as loss of air-flow in the ovens, excess solvent concentration, a high temperature alarm, or interruption of the light screen.

In the event of an emergency stop condition, the following reaction steps will take place.

1. All nip rolls will open
2. Drive roll will open
3. Backing and metering rolls will open
4. Heat to hotplates and ovens will be turned off and reset to 500°F maximum when restarted
5. Electrical power to motors will be shut off
6. Motor clutches will disengage
7. Rewind clutches will disengage
8. All sensors and instruments will remain operable and will continue to monitor the situation

The emergency stop condition will remain until the point of activation has been cleared and the reset buttons on the wall mounted control cabinet and the control console have been pressed.

In the event of a high solvent concentration alarm, loss of air flow in hotplate hoods or oven, or a high temperature alarm, the following reactions will take place:

1. Heat to hot plates and oven will be turned off and reset to 500°F maximum when the system is reset.
2. Electrical power to the motors will be shut off.

3. All sensors and instruments will remain operable and will continue to monitor the situation.

Again, the alarm condition will remain until the problem is corrected and the reset button on the control panel pushed.

## **5.2 Air Flow**

If the air flow through the ovens is adequate, the system is safe without the gas detection system. The gas detection system is a back-up to improper air flow in the oven or hot plate hoods. An air flow sensor is located in the duct connecting all three hoods together. If the blower is not on, or fails, the machine's heating elements will turn off until the problem is corrected. As a general rule, there should be 10,000 cubic feet per minute of air mixed with every gallon of evaporated solvent. Therefore, the amount of air flow required, based on line speed, percent solvent in solution, and resin content, needs to be calculated. The air flow in each oven must be adjusted accordingly. A velometer is required to measure the flow rates accurately. Each oven is equipped with a damper to adjust flow. The hot plates use air from the surrounding room as make up air to help keep the air flow to desired levels. The oven make-up air is supplied by the inlet blower. Appendix IV describes the necessary calculations to determine safety factors that ensure sufficient air flow is passing through the ovens.

## **5.3 Gas Detection**

With the machine's capabilities to process prepreg from solution, solvent vapor concentration must be detected to ensure a safe atmosphere and to prevent the possibility of explosion. Therefore, a real time gas detection system has been designed into the system. Samples are drawn from the three hot plate and oven assemblies using a venturi vacuum pump. The sample gas is detected by the combustible sensor which analyses the sample. The signal is sent from the sensor to the system module where the concentration is displayed. If the concentration of any of the channels exceeds 25% of the lower explosion limit (LEL) of the solvent being tested (when calibrated correctly), the system will shut off all heater and motor power on the machine.

With the number of different solvents in which the process may be exposed to, the operator must take great care in calibrating the gas detection system for the particular solvent being used. If the system is not calibrated correctly, the gas



detection system may become useless and may even result in serious injury to the operator. A list of solvents and their response to the system are listed in the operating manual. The system uses methane as the calibration standard. The instructions presented in the GasTech manual should be followed to calibrate the sensor. The conversion factor in the table should be used to calibrate the sensor for a specific gas other than methane.

For gases that are not in the table and are not available from GasTech, it is strongly recommended that the instrument be calibrated using a known concentration and its associated L.E.L. for the actual gas to be detected. Each channel should be calibrated any time the solvent is changed and there is the chance of an unsafe condition.

## **5.4 Oven Temperature Alarms**

Each electrically heated surface is equipped with an over-temperature circuit to avoid run-away conditions. The over-temperature alarms all fail in the safe mode (i.e. open circuits). If an alarm condition arises, an open circuit in any one of the controllers will cause an open circuit to the primary heater contractor coil, causing all heaters to shut down. The over-temperature condition will remain until the reset on the wall cabinet is pushed. When a Watlow 985 controller goes into an over-temperature condition (500°F), the controller has to be reset by pressing the 'manual' button on the controller twice to clear the alarm.

The alarms on the Watlow 985 temperature controllers are set at 500°F. This limit is lower than the auto-ignition temperature of most solvents that will be used. The operator should always know the solvents auto-ignition temperature before starting the machine. The 500°F limit can be overridden by pressing the over-ride button. It is intended that the over-ride only be used when processing solventless systems. If an alarm condition arises while the over-ride is activated, the system will reset to the 500°F degree limit.

The main over-temperature controls are located inside the wall cabinet and are set at 850°F, high enough to get up to the design allowable of 800°F. The reset button on the control cabinet must be pushed following power start-up to close the output contacts of these controllers.

## **5.5 Oven Protection**

The oven is equipped with a few safety mechanisms to protect the oven's longevity. The oven heater will not turn on unless the water pump switch on the

chiller and inlet blower are turned on. Water must be circulated into the cooling jacket of the infrared sensor to decrease the chances of damage due to excessive heat. Periodically, the flow valve for the sensor cooling water is checked and the pressure is adjusted to read between 12-15 psi.

The blower must be on to prevent the heating elements from overheating due to lack of air flow. The air flow across the heating coil is lower than the manufacturers recommendation, but with the heater sheath over-temperature protection the air flow will be adequate. To control the sheath temperature, an on/off controller is set for a maximum temperature of 900 °F. This temperature is low enough to prolong the life of the heater. The lower the sheath temperature the longer the heater will last. If the process can withstand it, a 700°F setpoint would be ideal.

## **5.6 Chain and Light Guards**

To facilitate the maintenance of the bearings, chains, brakes and clutches, a light curtain is used behind the machine to prevent contact with moving parts. All sprockets and chains are open in the back of the machine. The light curtain protects the entire length of the machine. If the light screen is interrupted by an object, the motors will stop and will not restart until the restart button is pushed. Indicator lights are on the body of the curtain and are on when the screens are operating correctly with no obstacles breaking the beam.

From the front of the machine, the chains are guarded by chain guards which prevent the operator from reaching the chain. All the guards can be easily removed if needed.

## **6.0 OPERATING PROCEDURE**

### **6.1 Initial Procedure Prior To Machine Operation**

The following outlines the steps that must be followed prior to operating the tape machine.

1. Depending on the resin system to be impregnated, take the necessary steps to confirm that the machine will be operated in a safe manner (Section 5).
2. Decide what the required prepreg specifications should be (Section 3).
3. Depending on how much fiber is required, calculate the desired number of fiber ends (Section 4.1).

4. Thread the comb in the appropriate manner as previously described (Section 4.1).
5. Check all the nip gaps are even from one side to the other (as discussed in Section 4.3)
6. Decide on how impregnation is to be performed.

Impregnation with precast films

This method is typically used for resin systems that process like conventional hot-melt thermosets and have viscosities in the 1000 centipoise range.

Solution dip-tank methods

Solution impregnation is used for polymer systems that have very high melt viscosities and can only be processed by dissolving in an appropriate solvent system.

Set the metering bar specifications

This is performed using feeler gages and two small wedge blocks that are located at either end of the bar.

Direct impregnation at the reverse roll coater

The solution dip-tank has an upper level viscosity range of approximately 25,000 cp. Some solutions may not be processable via the dip tank method as the solution is pulled away from the impregnation bars and, therefore, need to be impregnated at the reverse roll coater.

7. Arrange the desired paper set-up with paper and cores at the appropriate take off and take-up locations.

Numerous combinations of operating procedures are possible and the tape machine possesses a variety of locations to apply and take up the release papers. For example, in the drying study it was decided to remove both the papers at nip stations #2 and #3 to promote further drying in the main oven.

8. Attach all the fiber to the release paper and thread through the machine by pulling the paper and fibers through each module in sequence.
9. Set all hot plate temperatures.
10. Set the oven temperature.
11. Set the nip roller temperatures.
12. Keep the nips down in a preset open mode.

The correct start-up position for the nip rollers is to initially lower them and drive the wedge blocks completely inwards to obtain an open gap, this allows the prepreg web to pass freely.

13. Set the initial paper tensions.

These adjustments can be made by applying more pressure to the brakes at the individual paper unwind controls. These are located on the main control console.

14. Fill the dip tank (or the reverse roll coater reservoir for filming processes).

Do not fill completely. Air brought into the dip tank by the fibers may cause the liquid level to rise too high.

15. Set the initial running speed

## 6.2 Operation

1. Begin the prepregging operation.

During the operation there are a number of locations along the machine length that require attention. (A) The loose fibers that build around the comb must be continually removed to ensure that individual fiber tows are not broken. (B) The fiber spools on the creel must be checked to ensure that no tow has become trapped and caught in the spool holder bearings. (C) The dip tank needs to be continually monitored to ensure sufficient impregnation is taking place. (D) For solution impregnation the nip station #2 must be lowered as soon as the first solution impregnated tows pass through the hot plates. The gap closing must be optimized by tuning the machine at all nip stations that are being operated. It is important to note that the oven and roller temperatures will control the amount of resin flow at the nips. (E) The run sheet described in Table II should be accurately completed for all the parameters used in the operation.

(E) Monitor the film or prepreg specifications. Perform any necessary changes in the operating parameters that will provide material which meets the desired specifications. (G) Resin areal weights of films can be readily obtained by cutting a sample of known area and weighing the film sample. The fiber areal weights and resin solid contents of prepreps need to be obtained by a different method, especially if polyamid acid solutions are being used, where prepreg samples contain both residual solvent and water from the imidization process. The

percent volatiles and resin solids content of such samples may be obtained using the following methods.

#### Percent volatiles

W1 = Initial weight of the wet prepreg sample

W2 = Final weight of sample from B-staged material in an oven set to 500°F for half an hour.

$$\text{Percent volatiles} = \frac{W1 - W2}{W1} * 100$$

#### Resin Solid Content

Assuming fiber areal weight = 145 g/M<sup>2</sup> (=1.45 g/cm<sup>2</sup>)

$$\text{Resin solids \%} = \frac{W2 - 1.45}{W2} * 100$$

The above equation is for a sample with dimensions 10cm x 10cm.

$$\text{True fiber areal weight} = W4 = W3 * 100$$

Where W3 = Weight of fiber from a sample of known area that has been extracted and washed with solvent to obtain the carbon fibers which are then dried and weighed.

$$\text{True Resin Solid Content} = \frac{W2 - W4}{W2} * 100$$

The above methods should be performed on samples taken during the prepregging operation. Also, samples should be submitted for acid digestion studies to obtain a more rigorous and accurate analytical result.

## 7.0 DESIGN CORRELATIONS

Establishing the desired protocol for the prepregger requires an understanding into the relationship between machine operator variables and product requirements. As indicated in the prepregger operation matrix (Section 2.0), and in the discussions

of the operating window (Section 4.5), product quality requirements restrict operations to within an envelope of operating parameters.

During the initial operation of the machine, suitable conditions and processing parameters for prepregging a number of resin systems developed by NASA Langley were obtained. A variety of high performance prepreg material systems were produced on Hercules IM7 carbon fiber. These included LaRC™ RP46, a PMR type resin processed from a methanol solution and two polyamide acids, LaRC™ IA and LaRC™ ITPI prepregged from N-methyl pyrrolidinone (NMP) solution.

To fully utilize the machines ability, it is necessary to utilize each of the variables within each module. In this regard, an initial experimental plan has been outlined to obtain and understand the interrelation of various design variables and correlations.

## **7.1 Tape Machine Experiments**

A number of experiments are proposed following the Taguchi experimental design approach to be performed after the parameters leading to high quality LaRC™ ITPI prepreg have been obtained. These experiments focus on key components of the prepregger which dictate the quality of the resulting prepreg product.

Once the conditions for making quality prepreg have been determined, a series of deviations from these established operating parameters will be made to help define processing windows for each individual station. Initial experiments will focus on the stations listed below. After the completion of each excursion from the established operating conditions, all settings will be returned to the position for stable running conditions before starting the next series. In this way the boundaries of the operating window for making quality prepreg will be determined for various combinations of operating conditions.

### **STATIONS**

- |   |             |
|---|-------------|
| 1 | Hot-plate 1 |
| 2 | Hot-plate 2 |
| 3 | Oven Temp3  |
| 4 | Nip #2      |
| 5 | Nip #3      |
| 6 | Nip #4      |

The sequence of experimental parameter variations for finding the boundaries of the operation window will be:

### Nip Stations

- Expt. #1      Open nip station #4 until gaps appear.  
                  Monitor the gap width.  
                  Sample the prepreg to determine the percent solvent in prepreg.
- Expt. #2      Open nip station #3 until gaps appear.  
                  Monitor the gap width.  
                  Sample the prepreg to determine the percent solvent in prepreg.
- Expt. #3      Open nip station #2 until gaps appear.  
                  Monitor the gap width.  
                  Sample the prepreg to determine the percent solvent in prepreg.  
                  Monitor the observations at nips #3 and #4.

### Hot Plate Temperatures

- Expt. #4a      Increase temperature at 10°F intervals.  
                  Too much resin volatilization, creates higher resin viscosity.  
                  The prepreg is less compliant and gaps appear.  
                  Monitor gap sizes.  
                  Sample prepreg before nip #2 to find the percent volatiles.
- Expt. #4b      While at these higher temperatures, change the gaps to  
                  determine the required pressures needed for gap elimination.  
                  Prior to moving to a second temperature, change the gap to the  
                  original stable setting.
- Expt. #5      Decrease temperature at 10°F intervals.  
                  Lower devolatilization occurs and the resin viscosity decreases.  
                  The nips squeeze out too much resin as excessive pressure is

being applied and unacceptable resin wash out is observed. Also, the Fiber Areal Weight is no longer within specifications and the prepreg width is too large.

Monitor the percent volatiles to help determine percent solids and therefore obtain the viscosity of the resin.  
Measure the extent of the fiber movement and resin wash out per temperature decrease.

#### Oven Temperatures

Expt. #6      Increase the Oven temperature to obtain gaps.  
Find out if closing nip 4 will eliminate these gaps.  
Find out if heating nip 4 will eliminate these gaps.

The information obtained from these experiments will establish the range for each parameter within which acceptable prepreg can be made. This information will be incorporated in design and operating correlations for use in the design of future prepregging machines.

## **8.0 INITIAL RESULTS AND OBSERVATIONS**

From initial operations the following observations have been made that provide useful insight into the production of quality prepreg.

The metering equations based on densities that are used to calculate the metering bar gap dimensions are insufficient. Often the bar gap needs to be modified to allow the desired quantity of resin to be impregnated into the tow. Using the calculations described in section 3.3, it was found that the actual metering bar gap had to be significantly increased in order to obtain the desired solids content on the prepreg. For the LaRC™ ITPI polymer in NMP (30 percent solids solution) samples of prepreg were taken as the metering bar gap was opened in increments of 0.001 inches. The actual weight percent polymer on the prepreg was measured by performing acid digestion on the samples that had been B-staged for two hours at 500°F.



Figure 12 illustrates the discrepancy between the actual percent solids on prepreg and that predicted by the density equations presented in Section 3.3. One possible explanation, which deserves further study, is that the original equations, based on densities, do not apply as the prepreg web is not fully impregnated (in this case central regions of the fiber bundle are not fully wetted). The large amount of air bubbles observed within the dip tank suggest that entrapped air within the impregnation zone may be one reason why the original density equations are not followed. Analysis of this problem needs to be made. A possible starting point could be the calculation of the Prepreg Flow Number (PFN) (Section 3.4), of the impregnation zone within the dip tank for each solution studied.

Devolatilization of solvent is a difficult process. The temperatures required to remove high boiling solvents may advance the resin beyond an acceptable level, where its processability is diminished. In certain instances where polyamid acids were being used, the excessive heating may cause the imidization reaction to proceed. This may reduce the processability of the resulting polymer.

The removal of splits is a vital process in the production of quality prepreg. To date, it has been noted that the formation of gap-free prepreg at nip stations #2 and #3 is extremely important in producing quality product. When gap-free prepreg enters the oven, drying out additional solvent may lead to the formation of more gaps. However, the gap formation could be reduced by increasing the fiber areal weight at the dip tank. This insures that the collimated fiber tows are kept together during the hot plate drying process and are less likely to split when the prepreg web is dried further in the high temperature oven.

The volatiles can be removed quite readily by increasing the oven temperature. For example, an increase in oven temperature from 370 °F to 430 °F decreased the percent volatiles by approximately 6% (Figure 13). However, the quality of the resulting prepreg was found to deteriorate as the temperature increased and the gap formation became more pronounced.

In an attempt to remove these gaps the width of the dry web was decreased at the point of entry into the dip tank. The desired width could still be obtained by controlling the amount of squeeze at the second nip station. Although in practice this was found to difficult too perform. A decrease in the gap width tended to squeeze out excess resin rather than increase the width of the tow. That is, resin flows in the transverse direction more readily than the carbon fibers.

It was noted that the best quality prepreg, occurring at the end of the product line, was obtained when the prepreg web exiting from the #3 nip was completely gap

free. In these cases, further solvent removal tended to contract the web and few gaps were observed.

With regards to the prepreg uni-tape made from powder coated towpreg. The initial trials produced a product that had a thickness of 0.017 inches. This suggests that entrained air remains within the prepreg and that complete consolidation of the individual powder coated tows was not achieved with the hot sled. A thickness of 0.005-0.007 inches would represent a well consolidated uni-tape. The limiting factor in this process to date is the release material. A suitable material is needed to protect the prepreg product from the rollers within the hot sled. In the initial trials stainless steel was utilized, however the release of the hot product was insufficient and damage to the prepreg occurred. Stainless steel was replaced with Teflon® coated glass cloth, which worked well. A limiting factor using this material was the amount of pressure that could be applied. Any pressures above 1000 psi damaged the glass cloth and the polymer adhered to the hot rollers. Future work using this process will involve thick stainless steel or aluminum foil (approximately 0.005 inches in thickness), that has been coated with a high temperature release agent. Possible coatings include silicones and FEP's (fluorinated ethylene propylene copolymers).

## **9.0 CONCLUDING REMARKS**

This memorandum presents an introduction to the NASA multi-purpose prepregging unit which is now installed and fully operational at the Langley Research Center in the Polymeric Materials Branch. A description of the various impregnation methods that are available to the prepregger is given. Machine operating details and protocol are provided for its various modes of operation. These include, where appropriate, the related equations for predicting the desired prepreg specifications. Also, as the prepregger is modular in its construction, each individual section is described and discussed. Safety concerns are an important factor and a chapter has been included that highlights the major safety features.

A number of polymeric materials have been prepregged, these are discussed in individual sections relating to the various impregnation processes. Initial experiences and observations for fiber impregnation with these polymers are described. These first observations have given great insight into the areas of future work that need to be addressed. Future memorandums will focus on these individual processes and their related problems.

An appendix describes appropriate calculations that are related to the various processes. Also, as a complete engineering analysis needs to be performed on the prepregger, the appendix includes a series of analyses which relate to processes that form the various prepregging methods.

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## Appendix

### I Prepregging Calculations

#### Ia Prepregging Variable Definitions

FAW = Dry fiber areal weight

RAW = Resin areal weight

PAW = Dry prepreg areal weight

(PAW)<sub>wet</sub> = Prepreg areal weight with volatiles

W<sub>f</sub> = Fiber Areal weight fraction of PAW or (PAW)<sub>wet</sub>

W<sub>m</sub> = Matrix weight fraction of PAW or (PAW)<sub>wet</sub>

W<sub>solvent</sub> = Solvent weight fraction of (PAW)<sub>wet</sub>

W<sub>solid</sub> = Solid weight fraction of solution (i.e. 40% solid solution)

t = Dry prepreg thickness

(t)<sub>wet</sub> = Prepreg thickness with volatiles

ρ<sub>c</sub> = Prepreg density

(ρ<sub>c</sub>)<sub>wet</sub> = Prepreg density with volatiles

Ends = # of fiber spools

y = Fiber yield

w = Prepreg width

#### Ib Pre-Cast Film Calculations

The material balance calculations that follow are typical of those used to calculate the metering gap width for reverse roll coating to prepare films of a desired thickness to be used in the double sided coating impregnation process. These equations do not include the effects of shear thinning.

For hot-melt processable resins;

$$(t_{\text{film}}) = \frac{\text{RAW}}{2 (100)^2 (\rho_m)} \quad \text{I.1}$$

For filming resins from solution;

$$(t_{\text{film}})_{\text{WET}} = \frac{(\text{RAW})_{\text{wet}}}{2 (100)^2 (\rho_{\text{sol}})_{\text{wet}}} \quad \text{I.2}$$

where

$$\rho_{sol} = \frac{1}{\frac{w_{solid}}{\rho_m} + \frac{w_{solvent}}{\rho_{solvent}}} \quad 1.3$$

$$FAW = \frac{\# \text{ ENDS } \times 39.37 \times Y}{W} \quad 1.4$$

$$PAW = \frac{FAW}{1 - w_m} \quad 1.5$$

$$(PAW)_{wet} = FAW \left( \frac{w_m}{(1 - w_m) w_{solid}} + 1 \right) \quad 1.6$$

$$RAW = PAW - FAW \quad 1.7$$

$$(RAW)_{wet} = (PAW)_{wet} - FAW$$

#### EXAMPLE

$$FAW = 145 \text{ g/m}^2$$

$$w_m = 35\% \text{ or } 0.35$$

$$\rho_m = 1.25 \text{ g/m}^2$$

$$PAW = 145 \text{ g/m}^2 / (1 - 0.35) = 223 \text{ g/m}^2$$

$$RAW = 223 \text{ g/m}^2 - 145 \text{ g/m}^2 = 78 \text{ g/m}^2$$

$$t_{film} = (78 \text{ g/m}^2 / 2(100)^2 (1.25 \text{ g/m}^2)) = 0.00312 \text{ cm} \\ = 0.0012 \text{ in}$$

To compensate for the presently unquantifiable effect of shearing the resin during filming, the actual coater gap setting used is typically twice the value calculated using the above relationships. In this example, the actual gap setting would be about 0.0024 inches.

#### Ic Metering Bar Calculations for the Solution Dip-Tank Process

Assumptions included into the dip-pan metering are:

1. The specific gravity of the solutions follow the proportional rule of mixtures.

2. Resin solution fully penetrates the tow ensuring that no air is present within the fiber bundles.
3. The solutions exhibit Newtonian behavior during shear while they flow.

$$t_{wet} = \frac{(PAW)_{wet}}{(\rho_c)_{wet} (100)^2} \quad 1.8$$

The above equation does not apply if full impregnation is not observed. For example, in the worst case, when zero impregnation occurs into the fiber bundle, the metering bar gap size is increased as the  $(\rho_c)_{wet}$  term decreases.

Example for determining the metering bar gap dimensions

$$W_m = 35\%$$

$$FAW = 145 \text{ g/m}^2$$

$$\rho_f = 1.8 \text{ cm}^3$$

$$\rho_m = 1.25 \text{ cm}^3$$

$$\rho_{solvent} = 1.07 \text{ cm}^3$$

$$W_{solid} = 40\%$$

$$PAW = \frac{FAW}{1 - W_m} = \frac{145}{0.65} = 223.1 \text{ gM}^2 \quad 1.9$$

Assuming the solution fully penetrates the fiber bundle then;

$$(PAW)_{wet} = FAW \left( \frac{W_m}{(1 - W_m) W_{solid}} + 1 \right) \quad 1.10$$

$$= 145 \left( \frac{0.35}{(1 - 0.35) (0.40)} + 1 \right)$$

$$= 340 \text{ g/m}^2$$

$$W_f = \frac{FAW}{(PAW)_{wet}} = \frac{145}{340} = 0.426 \quad 1.11$$

$$RAW = PAW - FAW = 223.1 - 145 = 78.1 \text{ g/M}^2$$

$$w_m = \frac{RAW}{(PAW)_{wet}} = \frac{78.1}{340} = 0.230 \text{ g/M}^2 \quad 1.12$$

$$W_{\text{solvent}} = 1 - w_f - w_m = 1 - 0.426 - 0.230 = 0.344 \text{ gM}^2 \quad 1.13$$

$$(\rho_c)_{\text{wet}} = \frac{1}{\frac{w_f}{\rho_f} + \frac{w_m}{\rho_m} + \frac{w_{\text{solvent}}}{\rho_{\text{solvent}}}} \quad 1.14$$

$$(\rho_c)_{\text{wet}} = \frac{1}{\frac{0.426}{1.8} + \frac{0.230}{1.25} + \frac{0.344}{1.07}} = 1.459 \text{ gM}^2 \quad 1.15$$

$$t_{(\text{wet})} = \frac{(\text{PAW})_{\text{wet}}}{(\rho_c)_{\text{wet}} (100)^2} = \frac{340}{1.459 (100)^2} = 0.0233 \text{ cm} = 0.0092 \text{ inches} \quad 1.16$$

## Id Calculations for Volatiles Removed from Monomers

Adjustments to the  $W_m$  values are needed if monomers produce volatiles that add to the solvent volatiles during heating of the prepreg. These modifications are required when polymers such as PMR resins and polyamide acids are being prepregged. The following example illustrates how to estimate the percent volatiles that will be removed from the monomeric species from a given quantity of RP 46 (a PMR resin).

### Quantities of Monomer Used in Prepregging RP46 Resin

BTDE (Methyl ester of benzophenone tetra carboxylic dianhydride (BTDA)	=551.9g
3,4' ODA (4,4' Oxydianiline)	=209.5g
NE (Methyl ester of nadic anhydride)	= <u>134.4g</u>
TOTAL	= <u>895.7g</u>
Total weight percent solids	=69.2%
Total weight of solids	= <u>619.8g</u>

The strategy for the calculation is to determine the number of moles of methanol that are released from the methyl ester of BTDA and NE when they cyclize to form the anhydride. This quantity of volatiles should be added to the calculated amount of water that is produced from the imidization of the amide acids. This water is produced from the reaction between the amine and the anhydrides.



1. Calculating the methanol and water that are released from the BTDE.

Molar mass of BTDE.	=386 g/mole
Mass of BTDE solution being used.	=551.9g
Therefore, total amount of BTDE in 50% solution.	= 275.95g
Total molar amount of BTDE.	=0.7149 moles
Total molar amount of methanol being lost (2x 0.7149). (The BTDE is a diester).	=1.429 moles
Total weight of methanol being removed from BTDE.	=45.75g

There are two anhydride groups per molecule, each will react with an amine releasing water from the imidization reaction.

Total molar amount of water being evolved.	=1.429 moles
Total mass of water being removed.	=25.772g

Total mass of methanol and water being released.	= <u>71.472g</u>
--	------------------

A similar calculation can be performed for the methanol and water released from the nadic anhydride.

Molar mass of nadic ester.	=196g/mole
Total mass of nadic ester being used.	=134.3g
Total molar amount of nadic ester being used. (This ester is monofunctional).	=0.655 moles
Therefore total moles of methanol being used.	=0.655 moles
Total mass of methanol being used.	=21.92g
Total molar quantity of water being removed.	=0.655 moles
Total mass of water being removed.	=12.33g
Total mass of water and methanol lost from nadic ester.	=34.25g

Total amount of methanol and water being released from both the NE and BTDE.	=105.727g
---	-----------

Total percent volatiles being removed from this material

105.727/619.8

=17%

After determining 17% volatiles will be emitted, this number can be used to estimate the correct  $W_m$  value for the solution dip tank calculations. As  $W_m$  is the matrix weight fraction of PAW or  $(PAW)_{wet}$  the prepreg coming from the machine should take into account the 17% volatiles. The prepreg is two thirds fiber and one third resin and as one third of seventeen is approximately six percent, this value should be added to  $W_m$  for calculations as to the metering bar gap .

## II Solution Prepregging Devolatilization Analysis

Solvent removal involves heat transfer and diffusion, the variables employed in describing these phenomena are:

$T$  = Temperature

$k$  = Thermal conductivity

$Y$  = Distance into the web thickness

$h_c$  = Heat transfer coefficient of web to the air

$D$  = Diffusivity of solvent in the vapour phase

$C$  = Solvent concentration

$U$  = Production rate =  $dz/d\theta$

$z$  = Distance along the web surface

$\theta$  = Time

$\alpha_p$  = Thermal diffusivity in the paper

$\alpha_w$  = Thermal diffusivity of the prepreg web

$X$  = Through the thickness distance

$k_w$  = Thermal conductivity of the prepreg web

In figure 6, at high solvent contents the section AB represents the constant rate of solvent removal. At the free surface, mass transfer of solvent occurs through an evaporation process. The appropriate boundary conditions equate the rate at which heat is conducted to the surface to the rate at which heat is lost from the surface. The heat loss includes both convection and evaporation. Using  $\Delta H_v$  as the heat of vaporization the following expression describes the process represented by the line AB.

$$k \frac{\delta T}{\delta y} = h_c (T - T_a) + \Delta H_v \left( -D \frac{\delta c}{\delta y} \right) \quad \text{II.1}$$

Hot plate design and operation consideration needs to take into account the conduction processes from the metal surface to the paper and finally to the web.

As the level of the solvent table travels down into the web thickness, the curve BC is followed in Figure 6. The solvent is considered to be a thin film transported as a rigid body at a line speed 'U' along the plate surface. Ficks law holds in this falling rate period, and using diffusivity 'D' as a function of solvent concentration 'c' the diffusion equation takes the following form.

$$U \frac{\delta c}{\delta z} = \frac{\delta}{\delta y} \left( D \frac{\delta c}{\delta y} \right) \quad \text{II.2}$$

The above equation defines the falling rate zone BC in Figure 6, and depicts the diffusion and conduction controlled rate of solvent removal. The AB constant rate line in Figure 6 is representative of drying solvent from the web surface.

Fourier's 2nd law for heat conduction determines the heat transfer and resin profiles in the paper and wet web. Figure 7 illustrates temperature profiles in the paper and web as they pass through the module. The temperature profile depends on system properties and are obtained from the appropriate solutions of Fourier's law.

Initially,  $\theta = 0$ , the web is at ambient temperature as it enters the hot-plate. For the purpose of the analysis, it is assumed that the air and plate temperature are constant. As the web passes over the plate, its temperature increases from ambient and approaches the plate temperature. The following differential equations describe the change in temperature with respect to time for both the paper and prepreg web.

FOR THE PAPER

$$\frac{\partial T}{\partial \theta} = \alpha_p \frac{\partial^2 T}{\partial X^2} \quad \text{II.3}$$

FOR THE PREPREG WEB

$$\frac{\partial T}{\partial \theta} = \alpha_w \frac{\partial^2 T}{\partial X^2} \quad \text{II.4}$$

Where  $X$  = Thickness  
 $T$  = Temperature  
 $\theta$  = Time  
 $\alpha$  = (Thermal Conductivity/Density) X Heat Capacity ( $k/\rho.C_p$ )

#### BOUNDARY CONDITIONS

PAPER  $T(0,X) = T_a$  Initially  $\theta=0$   
 $T(\theta,0) = T_p$  At the plate surface,  $X=0$ ,  
the temperature is  $T_p$

PREPREG WEB  $T(0,X) = T_a$  Initially  $\theta=0$

$$k_w \frac{\partial T}{\partial X}(\theta, b) = h_c [T(\theta, b) - T_a] \quad \text{III.5}$$

At the web-air interface  $x=b$ . The conduction rate through the web equals the air turbulent heat transfer rate.

#### PAPER AND WEB

$$k_p \frac{\partial T}{\partial X}(\theta, a) = k_w \frac{\partial T}{\partial X}(\theta, a) \quad \text{III.6}$$

At the paper-web interface, heat conducted from the paper equals heat conducted to the web.

These relationships describe heat transfer to the web as it passes over the hot plate. The transfer is by unsteady state conduction through the paper and into the web. Solutions to these equations would give the temperature and the rate of heating as a function of the exposure time.

### III Nip Station Pressure Analysis

#### IIIa Roller Footprint and Pressure

Variables employed in describing the phenomena associated with the roller footprint pressure are:

$P$  = Pressure

$\mu$  = Viscosity

$h$  = Web thickness at any value of  $x$

$y$  = Distance through the thickness

$x$  = Distance along the coating

$U_x$  = Velocity in the  $x$  direction

$R$  = Roller radius

$H_0, h_0$  = Initial web thickness

$U$  = Production Speed

$\bar{h}(\tau)$  = Average value of the web thickness

$W$  = Roller width

$Q$  = Volumetric flow rate

$F$  = Pulling force

$K$  and  $n$  = Rheological parameters (for a Newtonian fluid  $n=1$ )

$\tau_{yy}$  =  $yy$  component of shear stress tensor

$\tau_{xx}$  =  $xx$  component of shear stress tensor

$\tau$  = Shear stress tensor

$\Lambda$  = Rate of deformation tensor

$\int (n)$  = Integral of the pressure distribution.

The following analysis discusses the top half of the double roller system as the bottom half is symmetric to it, see the corresponding diagram in Figure 8.

The roller footprint,  $2L_s$  depends upon the roller diameter and the initial and final thickness of the compressed prepreg. Ardichivili (5) and more recently Gaskell (6) developed equations for the length,  $L$ , of the roller footprint from the point of initial contact to the nip. Middleman (1) has also a detailed description of the calendering processes. As indicated in the figure:  $h_0$  is the thickness at the initial point of contact,  $h_n$  is the nip thickness,  $R$  the roller radius, and  $U$  the linear speed. For the right triangle with sides  $R$  and  $L$  and hypotenuse approximated by  $R + h$ ,

$$(R + \Delta h)^2 = R^2 + L^2 \text{ assuming that } (\Delta h)^2 \ll 2(\Delta h)R \text{ and } L^2$$

$$L = [(h_o - h_n) 2R]^{1/2} \text{ or } h_o = h_n + \frac{L^2}{2R}$$

Using this relationship the web thickness,  $2h$ , varies along the footprint length,  $x$ , as follows:

$$h = h_n = \frac{(L-X)^2}{2R}$$

This expression for the thickness as a function of distance has been used in resin flow models to calculate the pressure profile as the material passes through the nip region.

As shown in Figures 9a, 9b, and 9c the top nip roller possesses a pneumatic pressure bearing system, with the bottom roller being fixed. The top roller bearing may be adjusted at various elevations using a screw drive employing "V" shaped wedge blocks. When the top roller is completely lowered (the "V" blocks are entirely open or pulled out) the top roller presses onto the web and the applied load is delivered by the pneumatic cylinders. The cylinders are 5 inches in diameter and the pressures are adjusted on the top of the pistons by the air supply.

For example, when the applied pressure equals 40 psi (and the wedge blocks are completely removed) as the diameter of each piston is 5 inches, the surface area of the piston over which the load is being applied is;

$$\frac{\pi}{4} (5^2) = 19.625 \text{ inches}^2$$

Hence the applied load exerted by each piston is

$$F = P \times A = 40\text{Psi} \times 19.625 = 785 \text{ lb's}$$

The pressure on the web is determined by knowing the area that the roller imprint makes in the prepreg as it passes through the nips and the applied pneumatic load (calculated above).

**N.B.** This method of determining the applied pressure can only be calculated when the gap is completely closed and the wedge blocks are completely removed.

When the blocks are completely pushed forward and lift the top roller, a gap exists between the two rollers. This provides a dam effect for the free liquid on the wet web. Liquid flows both vertically through the web and transversely out to the web edges and onto the paper. Under these conditions there is only a small hydrostatic pressure on the prepreg surface resulting from the partially closed rollers. This phenomena is similar to that at the metering bars.

A typical example of the necessary calculations to determine the roller footprint area are as follows.

The area of the nip roller footprint equals the width of the roller  $\times 2L$ . As an example, for an 8 inch diameter roller with  $h_0 = 0.02$  inches and  $h_n = 0.01$  inches

$$L = [(0.020 - 0.010) \times 2 \times 8]^{1/2}$$

$$= 0.4 \text{ inches}$$

The final footprint area for a 6 inch wide prepreg web

$$= 6 \times 2 \times 0.4 = 4.8 \text{ (inches)}^2$$

N.B. The two is used as the length 'L' is half the footprint length.

With 785 pounds of load from each piston the average pressure at each nip station  
 $= (2 \times 785 / 4.8) = 327 \text{ Psi}$

### IIIb Newtonian Coating

When a Newtonian fluid is being coated in an isothermal steady state operation, assuming that viscous forces dominate inertial forces and that the lubrication approximations are valid, the dynamic equations reduce to

$$0 = \frac{\partial p}{\partial x} + \mu \frac{\partial^2 u_x}{\partial y^2} \quad \text{III.1}$$

Using the following dimensionless variables with  $H_0$  being equal to the nip thickness during the calendering operation,  $U$  is the line speed,  $R$  is the roller radius,  $p$  is the pressure between the rolls and  $\mu$  is the material viscosity.

$$\zeta = \frac{x}{(RH_0)^{1/2}} \quad \eta = \frac{y}{H_0} \quad u = \frac{u_x}{U} \quad P = \frac{\rho H_0^3}{\mu U R^{1/2}}$$

and as in the calender model, assuming  $H_0/R \ll 1$

$$\frac{h(x)}{H_0} \equiv \tilde{h}(\zeta) = 1 + \frac{1}{2}\zeta^2 \quad \text{III.2}$$

The velocity field is (1):

$$u = 1 + \frac{1}{2} \frac{dP}{d\zeta} (\eta^2 - \tilde{h} \eta) \quad \text{III.3}$$

which gives  $u$  explicitly as a function of  $\eta$  and implicitly as a function of  $\zeta$  through the functions  $\tilde{h}(\zeta)$  and  $P(\zeta)$ .

If it is assumed that the liquid splits evenly to coat both the roller and the sheet, then the volumetric flow rate, the coating thickness, and  $\lambda$  are related by

$$Q = 2UH \quad \text{III.4}$$

$$\text{and} \quad \lambda = \frac{2H}{H_0} \quad \text{III.5}$$

from which

$$\tilde{h}_1 = 1 + \frac{1}{2}\zeta_1^2 = 3\lambda \quad \text{III.6}$$

and the pressure at the separation point is:

$$P(\zeta) = \frac{(6 - \frac{9}{2}\lambda)\zeta}{1 + \frac{1}{2}\zeta^2} - \frac{3\lambda\zeta}{\left(1 + \frac{1}{2}\zeta^2\right)^2} + \left(\frac{12}{\sqrt{2}} - \frac{9\lambda}{\sqrt{2}}\right) \tan^{-1} \frac{\zeta}{\sqrt{2}} + \frac{6\pi}{\sqrt{2}} \left(1 - \frac{3\lambda}{4}\right) \quad \text{III.7}$$

If the film splits at the point where  $u=0$  and  $p=0$ .



$$\lambda_0 = 1.30 \quad \text{III.8}$$

Once  $\lambda$  is known, the pressure distribution follows from equation III.7. The velocity field is then calculated from equation III.5. Stresses in the system may be calculated utilizing this information, when these are obtained one may find the roll sheet-separating force and the forces (and hence the power) required to turn the roll and pull the sheet. The roll/sheet-separating force, for example, is given by

$$F = W \int_{-}^{x_1} p(x) dx = \frac{\mu URW}{H_0} \int_{-}^{\zeta_1} P(\zeta) d\zeta \quad \text{III.9}$$

and for  $\lambda_0 = 1.3$

$$2.6 = \frac{FH_0}{\mu URW} \quad \text{III.10}$$

Once the pulling force is known it is possible to calculate the tensile stress exerted on the sheet. If this stress is high enough, it is possible that the sheet may undergo tensile deformation during coating with a consequent change in thickness. If a coating is laid down on such a deformed sheet, and if strain relaxation occurs subsequently, it is quite possible that the coating may "buckle" and become uneven. Thus a model for the tensile forces in the sheet itself could be a significant part of a complete coating analysis.

### IIIc Application of Power Law Fluids to Coating Processes

The power law case is treated in a manner that parallels the Newtonian model. Consistent with the lubrication approximation the power law is written as

$$\tau_{xy} = K \left[ \left( \frac{\partial U_x}{\partial y} \right)^2 \right]^{(n-1)/2} \frac{\partial U_x}{\partial y} \quad \text{III.11}$$

To avoid taking the fractional root of a negative quantity the velocity gradient is squared before the indicated fractional power is taken. When nondimensionalised the dynamic equations take the form

$$0 = -\frac{\partial P}{\partial \zeta} + \frac{\partial}{\partial \eta} \left\{ \left[ \left( \frac{\partial U}{\partial \eta} \right)^2 \right]^{(n-1)/2} \frac{\partial U}{\partial \eta} \right\} \quad \text{III.12}$$

where the dimensionless variables are defined as in the Newtonian case, except that

$$P = \left( \frac{H_0}{U} \right)^n \left( \frac{H_0}{R} \right)^{1/2} \frac{p}{K} \quad \text{III.13}$$

The solution proceeds as in the newtonian case. The key result, the pressure distribution, is given by

$$P = A \int_{-\infty}^{\zeta} \frac{[(\tilde{h} - \lambda)^2]^{(n-1)/2} (\tilde{h} - \lambda)}{\tilde{h}^{1+2n}} d\zeta \quad \text{III.14}$$

where

$$A = \left( \frac{1+2n}{n} \right)^n 2^{1/n}$$

Setting  $u(\zeta_1, 1/2h) = 0$  gives

$$\zeta_1 = \left[ 2 \left( \frac{2n+1}{n} \lambda - 1 \right) \right]^{1/2} \quad \text{III.15}$$

Then the condition  $P(\zeta_1) = 0$  gives (setting the upper limit at  $\zeta_1$  in equation III.23) an integral equation for  $\lambda$  which must be solved by trial and error. The calculations indicate that  $\lambda$  is increased by non-newtonian shear behavior but the effect is modest. Figure III.1 shows  $\lambda(n)$ .

The effect of  $n$  on the pressure distribution is significant, and figure Figure III.2 compares pressure profiles for several  $n$  values, including the newtonian case  $n=1$ . The roll separating force  $F$  may be calculated from the pressure distribution and written in the form

$$\frac{F}{W} = K \left( \frac{U}{H_0} \right)^n R f(n) \quad \text{III.16}$$

Figure III.3 shows  $F(n)$ .

**Figure III.1** Nondimensional coating thickness  $\lambda$  as a function of  $n$ .

**Figure III.2** Pressure profiles for newtonian and power law fluids

Figure III.3 Nondimensional roll-separating force as a function of  $n$ .

### III.d Viscoelastic Effects in Film Coating

If a viscoelastic fluid is subjected to the flow under consideration here, three phenomena might occur which would have to be accounted for in the model.

1. Nonnewtonian shear behavior
2. Normal stress development
3. Stress growth and relaxation due to the lagrangian unsteady nature of the kinematics

Viscoelasticity may appear explicitly in the constitutive equation for the fluid. With a model of the form

$$\tau + \theta_R \frac{\mathcal{D}\tau}{\mathcal{D}t} = \eta_o \Delta \quad \text{III.17}$$

where  $\theta_R$  is a characteristic relaxation time of the fluid, viscoelasticity has been incorporated into the formulation of the problem through the time derivative  $\mathcal{D}/\mathcal{D}t$ . For the shear stress  $\tau_{xy}$  then

$$\tau_{xy} + \theta_R \left[ u_x \frac{\partial \tau_{xy}}{\partial x} + u_y \frac{\partial \tau_{xy}}{\partial y} - \frac{1}{2} \left( \frac{\partial u_x}{\partial y} - \frac{\partial u_y}{\partial x} \right) (\tau_{yy} - \tau_{xx}) \right] = \eta_o \left( \frac{\partial u_x}{\partial y} - \frac{\partial u_y}{\partial x} \right) \quad \text{III. 18}$$

Now, if the lubrication approximations, namely,  $u_x \gg u_y$  and  $\delta/\delta x \ll \delta/\delta y$  are applied to the terms of equation III.18

$$\tau_{xy} + \theta_R \left[ \left( \frac{\partial u_x}{\partial y} \right) (\tau_{yy} - \tau_{xx}) \right] = \eta_o \frac{\partial u_x}{\partial y} \quad \text{III.19}$$

and

$$\tau_{xx} = \theta_R \tau_{xx} \frac{\partial u_x}{\partial y} \quad \text{III.20}$$

$$\tau_{yy} = -\tau_{xx} \quad \text{III.21}$$

Then

$$\tau_{xy} = \frac{\eta_o \left( \frac{\partial u_x}{\partial y} \right)}{1 + \theta_R^2 \left( \frac{\partial u_x}{\partial y} \right)^2} \quad \text{III.22}$$

and

$$\tau_{xy} = -\theta_R \eta_o \frac{\left( \frac{\partial u_x}{\partial y} \right)^2}{1 + \theta_R^2 \left( \frac{\partial u_x}{\partial y} \right)^2} \quad \text{III.23}$$

Equations III.22 and III.23 are identical to steady simple shear flow. Hence the lubrication approximations remove those terms of the constitutive equation associated with the lagrangian unsteady character of the flow.

The normal stresses enter the model in the calculation of the roll separating force. For a fixed nip separation as is the case for this NASA prerpegger when the wedge blocks control the gap:

$$\frac{F}{W} = \int_{-}^{+} [-T_{yy}]_{y=h(x)} dx = \int_{-}^{+} [p - \tau_{xx}]_{y=h(x)} dx \quad \text{III.24}$$

If the nip separation is not fixed but the force  $F$  is held constant, as in the case when the wedge blocks at the nip rollers are completely removed, then the normal

stress will affect the nip separation which will, in turn, affect the coating thickness. The effect of non-newtonian shear behavior on  $p$  must be examined before concluding anything about the change in total stress  $T_{yy}$ , which governs  $F$ .

#### IV Calculation and Procedure for Determining the Safety Factor when Operating the Tape Machine Ovens

The following equations were used to calculate the required ventilation when using a particular solvent during operation of the tape machine. N-methyl pyrrolidinone, NMP, is typically used in most of the solution prepregging runs and is utilized in the following example. The equations were taken from the 1989 National Fire codes on Ovens and Furnaces section 5-2. In these continuous processes, the safety ventilation rate is designed to prevent the vapor concentration in the oven from exceeding 25 percent of the lower explosive limit. The calculation below includes a safety factor of 4 in the equation.

$$\text{Required Ventilation} = \frac{444 \times \text{SpGr} \times (100 - \text{LEL})}{\text{VD} \times \text{LEL}} \quad \begin{array}{l} \text{Cubic feet of air referred to } 70^{\circ}\text{F} \\ (21^{\circ}\text{C}) \text{ per gallon solvent} \\ \text{evaporated, where:} \end{array}$$

Sp.Gr. = Specific gravity of solvent (water = 1)

VD = Vapor density of solvent vapor (air = 1)

LEL = Lower explosive limit expressed in percent by volume.

In SI units

$$\text{Required Ventilation} = \frac{3.33 \times \text{SpGr} \times (100 - \text{LEL})}{\text{VD} \times \text{LEL}} \quad \text{M}^3 \text{ of air.}$$

The flow rate into the oven must be calculated to maintain this required ventilation. The flow rate is in units of volume per minute and hence, depending on the tape machine running conditions, the amount of solvent evaporated will vary depending upon running speeds and solution concentrations.

Total volume of air required	Volume of air required per liter	Liters of solvent per minute entering
---------------------------------	-------------------------------------	--

for ventilation = of solvent X the oven  
(Flow rate)

To calculate the liters of solvent entering the oven requires a number of operating parameters to be determined. Also, the solvent weight loss per unit area, product width and line speed are required to calculate the amount of solvent evaporating within the oven. The following parameters are needed to calculate the safety factor at which the tape machine is operating. Typical values used in the following example are given with the individual parameters.

Run speed (1-20 ft/min)

Solvent concentration in the web ( $w_s = 30\%$ )

(Assume here that all the solvent is to be evaporated in one oven)

Fiber Areal weight (FAW =  $145 \text{ g/M}^2$ )

Desired resin solids on the prepreg ( $w_m 40\%$ )

Prepreg width

Solvent specific gravity

The following list details the procedure for calculating the safety factors;

1. Calculate the dry Prepreg Areal Weight (PAW).

$$\text{PAW} = \frac{\text{FAW}}{1 - w_m} = \frac{145}{1 - 0.4} = 242 \text{ g/M}^2$$

2. Calculate the dry Resin Areal Weight (RAW).

$$(\text{RAW})_{\text{dry}} = \text{PAW} - \text{FAW} = 242 - 145 = 97 \text{ g/M}^2$$

3. Calculate the solvent weight per square meter (S)

$$S = \frac{(\text{RAW})_{\text{dry}}}{w_s} = \frac{97}{0.3} = 323 \text{ g/M}^2$$

$$\text{convert to LB/ft}^2 = 0.66 \text{ LB/ft}^2$$

Convert to LB/ft . inch to bring in the prepreg width variable

$$S = 0.066 \text{ LB/ft}^2 \times 0.0833 \text{ ft/inch} = 0.0055 \text{ LB/ft . inch}$$

for one inch wide tape and using NMP solvent

4. Calculate the Evaporation Rate (ER) = S x W x V

S = Solvent weight per unit area (LB/ft . inch)

W = Prepreg width (inches)

V = Production rate (ft/min)

$$(ER = 0.0055 \times W \times V)$$

The evaporation rate is dependent upon the prepreg specifications, the product width and the production speed. It therefore varies for each prepregging run.

Table I describes how this evaporation rate varies with different prepreg widths and line speeds.

5. Determine the required air flow from the National Fire Code regulations (shown above).

$$\text{Required Ventilation} = \frac{444 \times \text{SpGr} \times (100 - \text{LEL})}{\text{VD} \times \text{LEL}}$$

Example for NMP (N-methyl-pyrrolidinone).

SpGr = Specific Gravity = 1.027 (G.A.F. MSDS sheet).

LEL = Lower explosion limit = 1.3 volume % (G.A.F. MSDS sheet).

VD = Vapor density = 3.4 (pg. 72 of National Fire Code 325M, 1984).

$$\text{Required Ventilation} = \frac{444 \times 1.027 \times (100 - 1.3)}{3.4 \times 1.3} = 10182 \frac{\text{ft}^3}{\text{gal}}$$

Converting to ft<sup>3</sup>/LB = 1189

6. Using temeptrature in degrees Rankine determine the required air flow for certain temperatures.



Using the above equations calculated at 70°F, the required air flow of 100°F air is given by;

$$\frac{100 + 459.7}{70 + 459.7} (1189) = 1256 \frac{\text{ft}^3}{\text{lb}}$$

7. Calculate the desired flow rate = Evaporation rate x required air flow.

The table describes the desired air flow of 100°F air for various prepreg widths and production speeds.

8. Calculate the safety factor =  $\frac{\text{Actual measured flow rate}}{\text{Desired flow rate}} \times 4$

Measured flow rate at the ovens is 200 ft<sup>3</sup>/min.

(NB. the factor of 4 is included as the given LEL volume is stated at 25%LEL)

Table I lists various evaporation rates for NMP and their corresponding desired flow rates. These values were obtained for prepreg products of a particular width and made using a set production rate. The calculated safety factor is also listed.

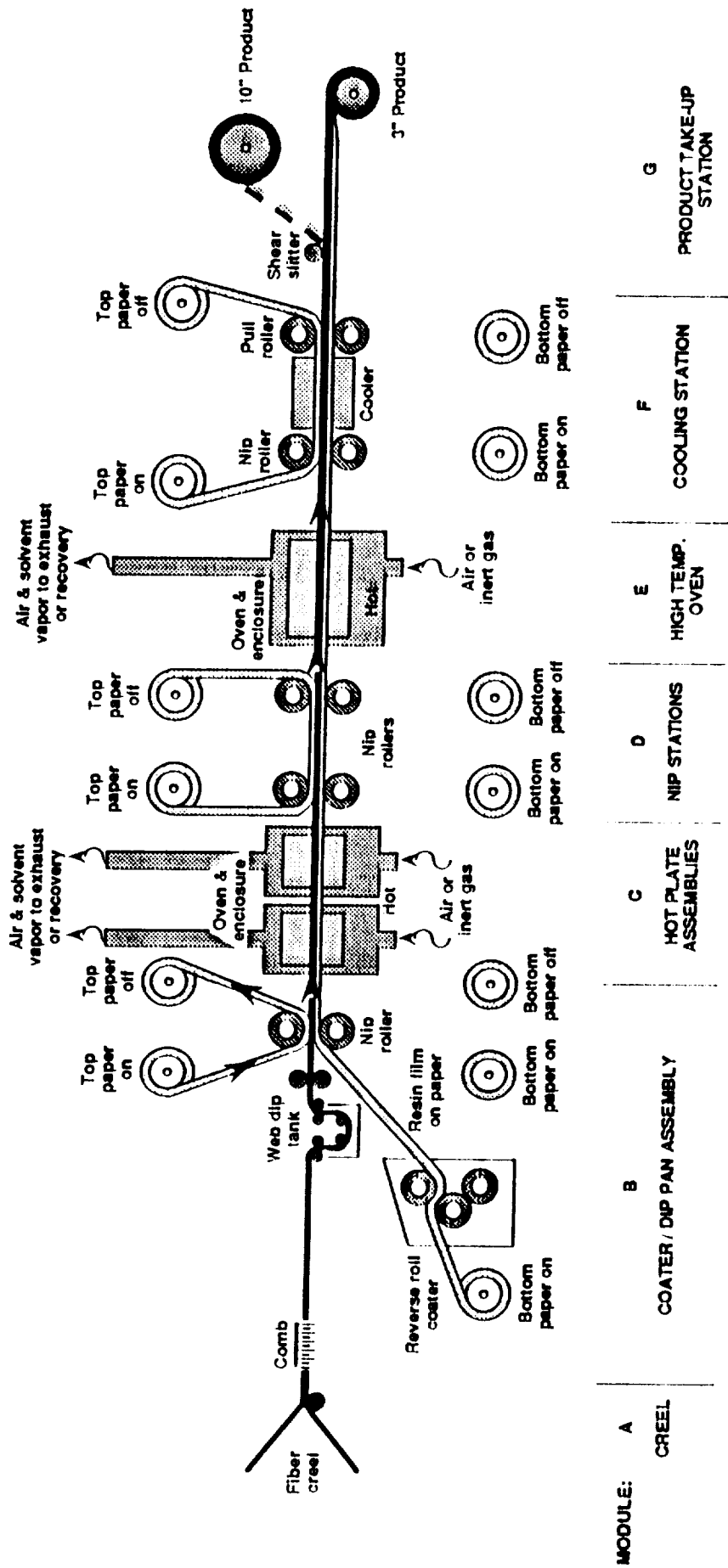
## 11.0 TABLES

**Table I-Safety Factor As A Function Of Line Speed And Prepreg Width**

Prepreg Width (inches)	Production Rate(ft/min)	Evaporation Rate (LB/min)	Desired Flow Rate (ft <sup>3</sup> /min)	Calculated Safety Factor
1	1	0.005	6.9	115
3	1	0.016	20.7	38.6
6	1	0.033	41.6	19.2
9	1	0.049	62.3	12.8
12	1	0.066	83.0	9.6
1	3	0.016	20.7	38.6
3	3	0.049	62.2	12.9
6	3	0.099	124.3	6.4
9	3	0.148	186.5	4.2
12	3	0.198	248.7	3.2
1	5	0.027	34.6	23.1
3	5	0.08	103.6	7.7
6	5	0.17	207.6	3.9
9	5	0.25	310.9	2.6
12	5	0.33	414.5	1.9
1	10	0.055	69.1	11.6
3	10	0.165	207.2	3.9
6	10	0.33	414.5	1.9
9	10	0.49	615.4	1.3
12	10	0.66	829.0	1.0
1	20	0.11	138.2	5.8
3	20	0.33	414.5	1.9
6	20	0.66	828.9	1.0
9	20	0.99	1243.4	0.6
12	20	1.32	1657.9	0.5

**Table II**  
**Machine Operating Parameters**

<b>Prepreg Batch Number</b> _____
<b>Coating Method</b> _____
<b>Fiber</b> _____
<b>Matrix</b> _____
<b>% Solids</b> _____
<b>FAW</b> _____
<b>Width (cm)</b> _____
<b>Fiber Yield (g/m<sup>2</sup>)</b> _____
<b>Dry Resin Content</b> _____
<b>Number of Tows</b> _____
<b>Comb #/Angle</b> _____
<b>Prepreg Thickness (cm)</b> _____
<b>Line Speed (M/sec)</b> _____
<b>Dip Tank Metering Gap</b> _____
<b>Nip Roll Data (Temp/Pressure/Gap/Speed Ratio)</b>
<b>Hot Plate Temperatures (°C)</b> _____
<b>Oven Temperatures (°C)</b> _____
<b>Unwind Tension Settings</b> _____
<b>Rewind Pressures</b> _____
<b>Product Pressures</b> _____



**Figure 1**  
Schematic Diagram Of The Tape Machine Modular Components

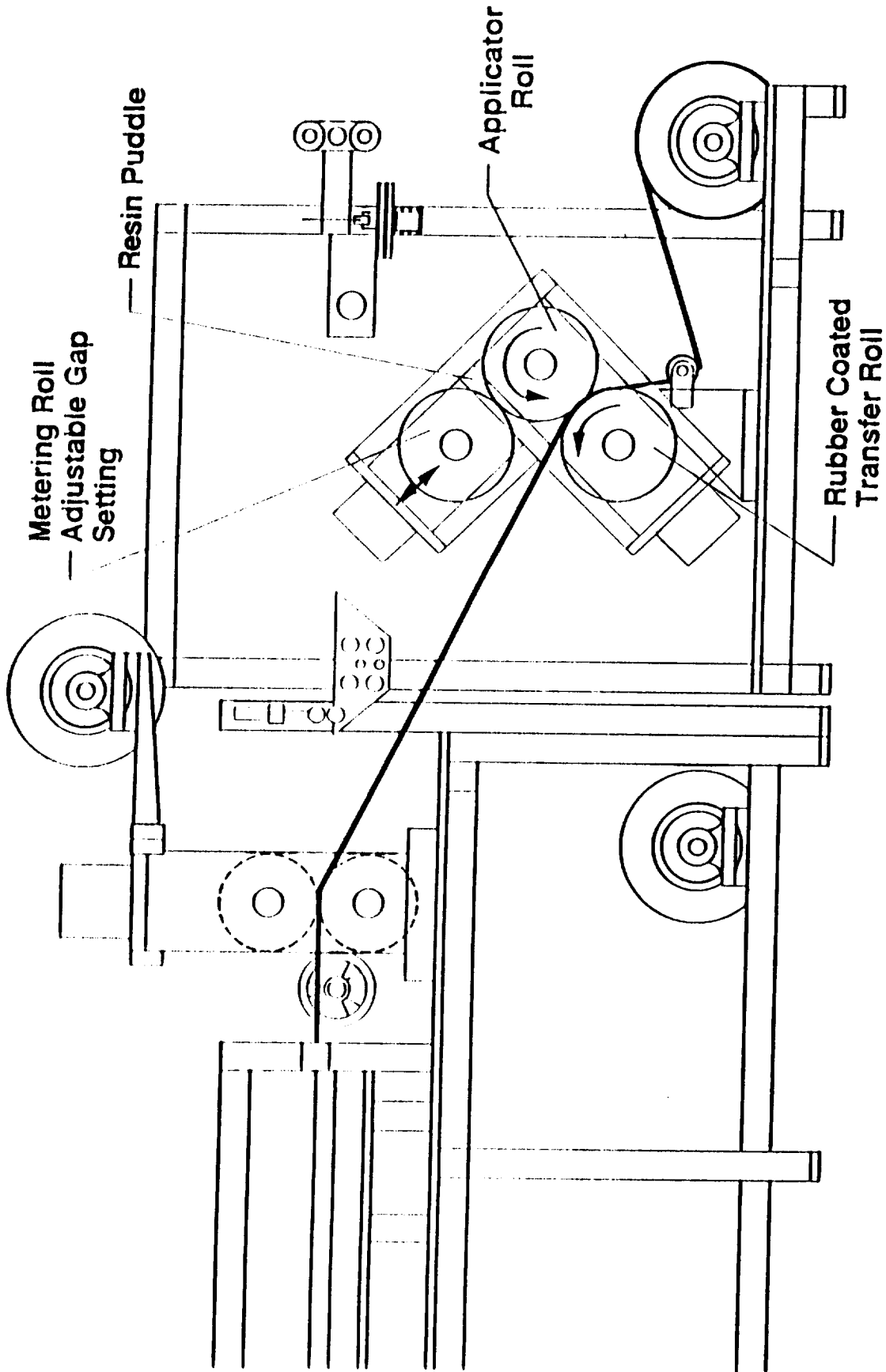


Figure 2. The Film Coating Process Using the Reverse Roll Coater.

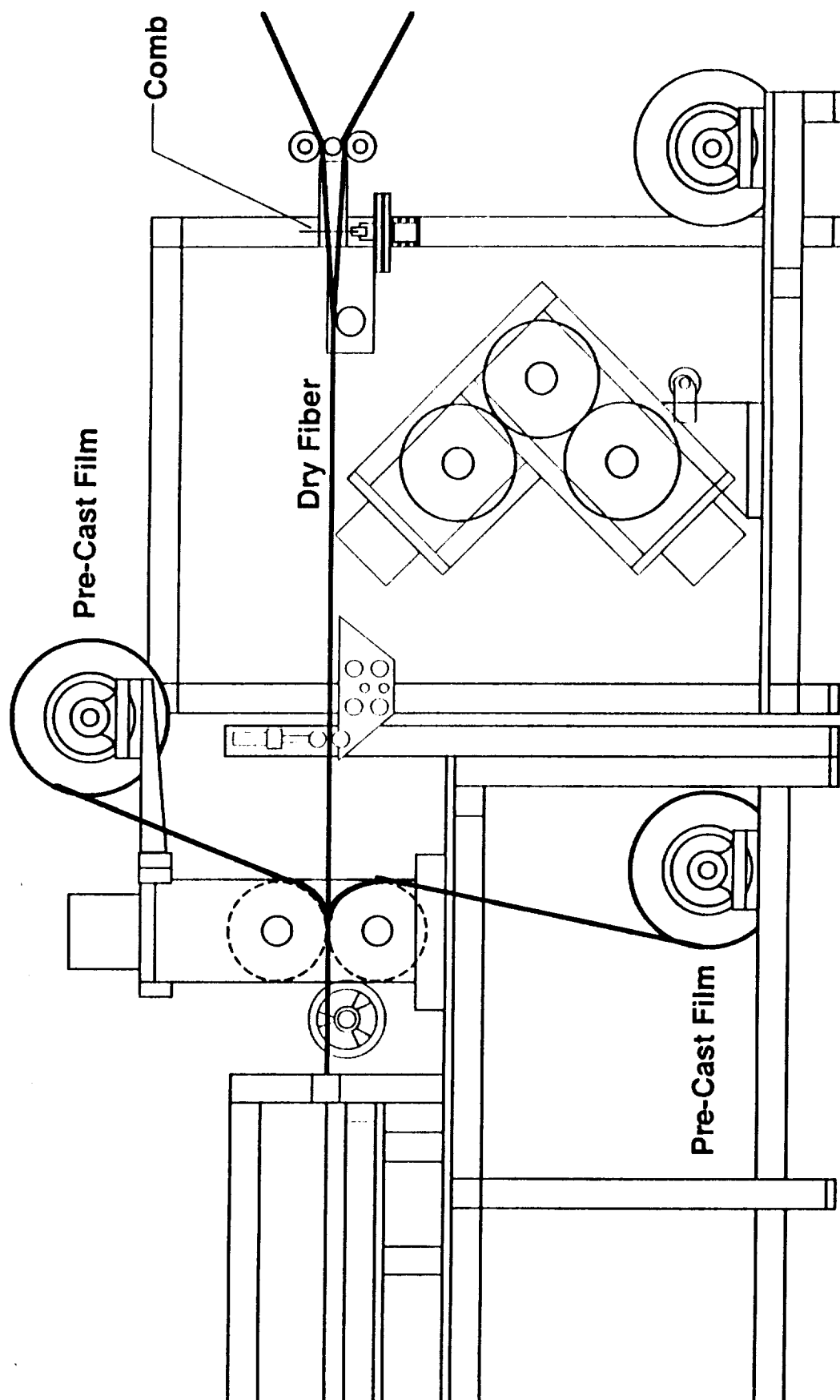


Figure 3. Common Fiber Impregnation Process Using Pre-Cast Films.

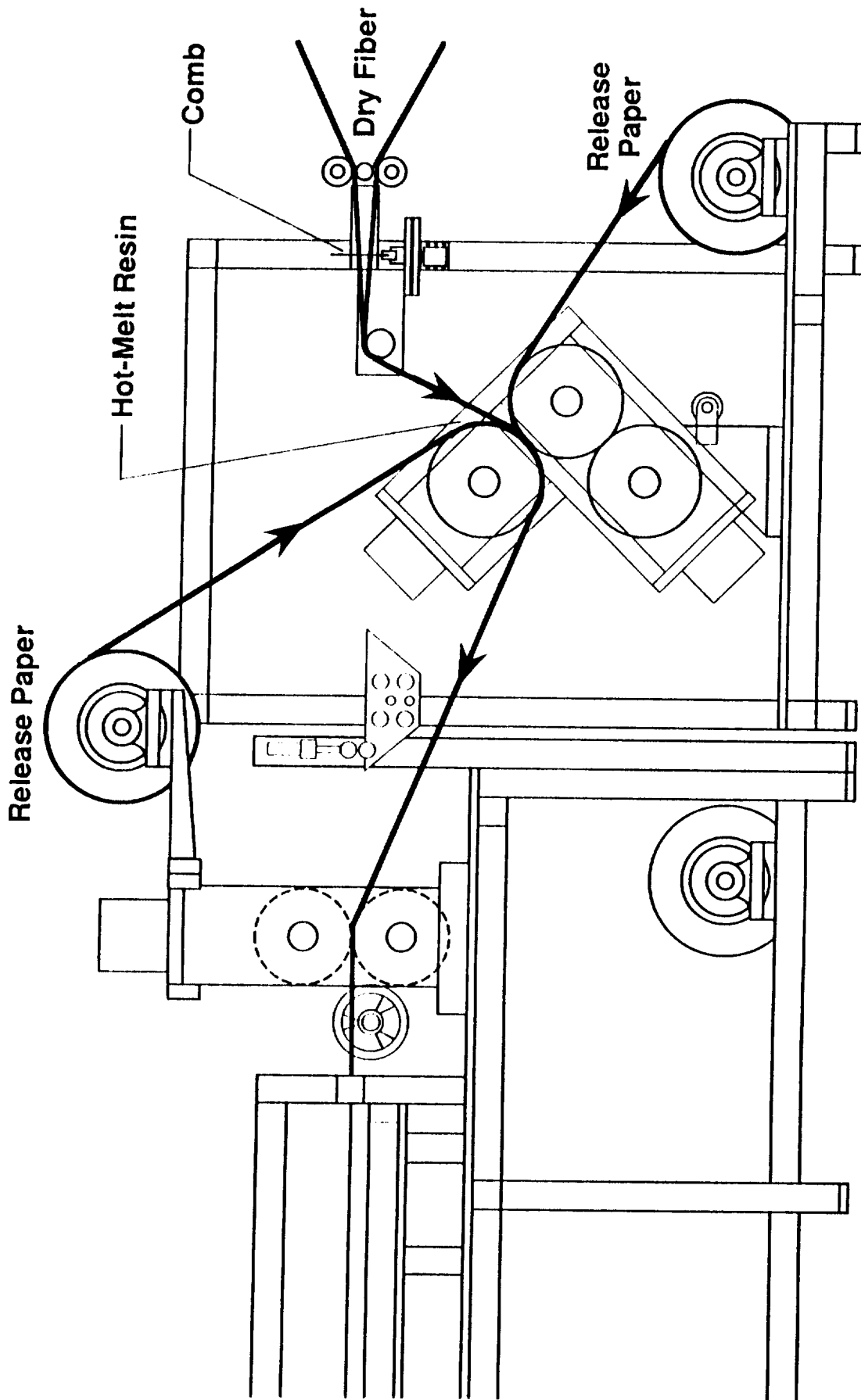


Figure 4. Direct Impregnation at the Reverse Roll Coater.

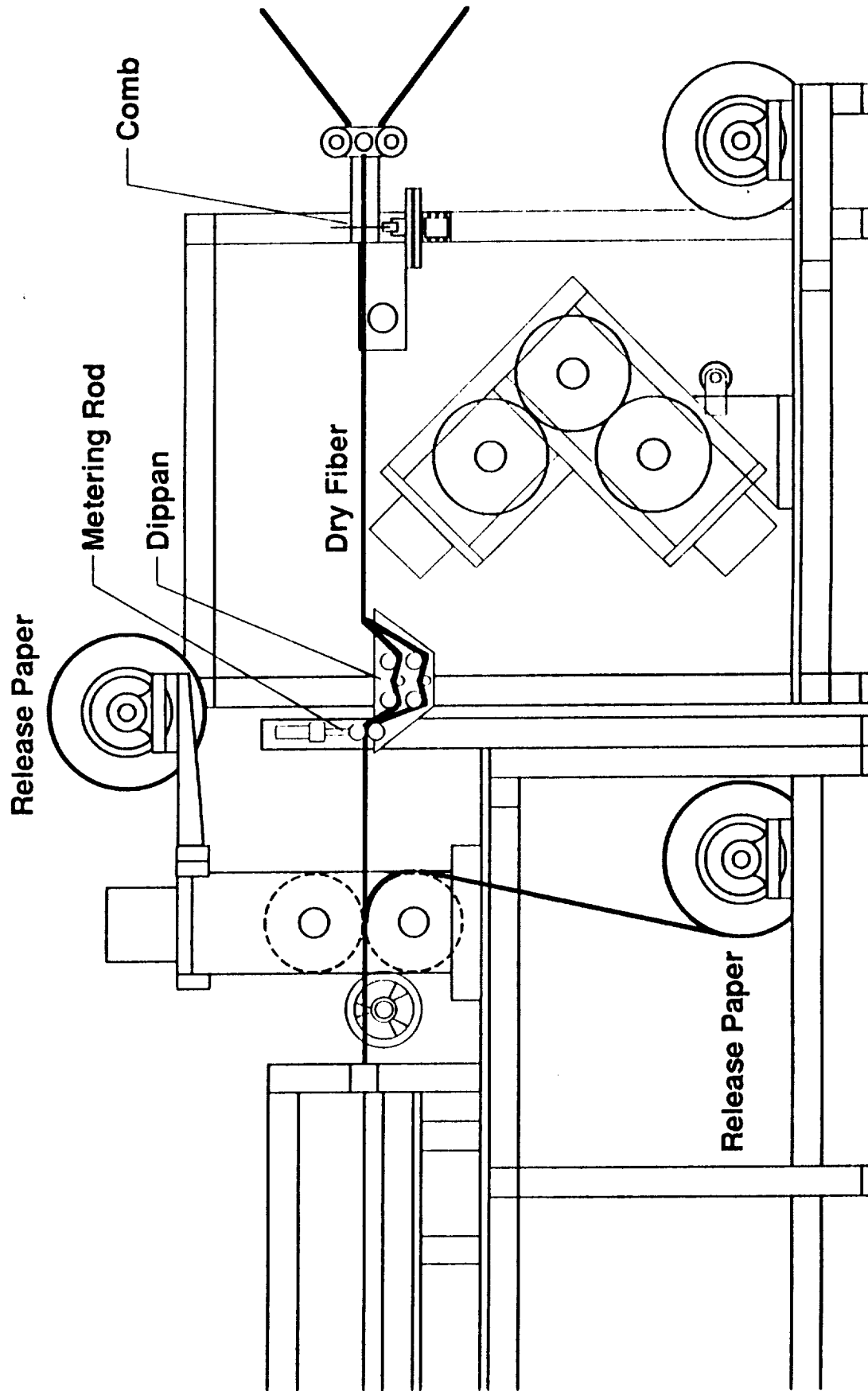
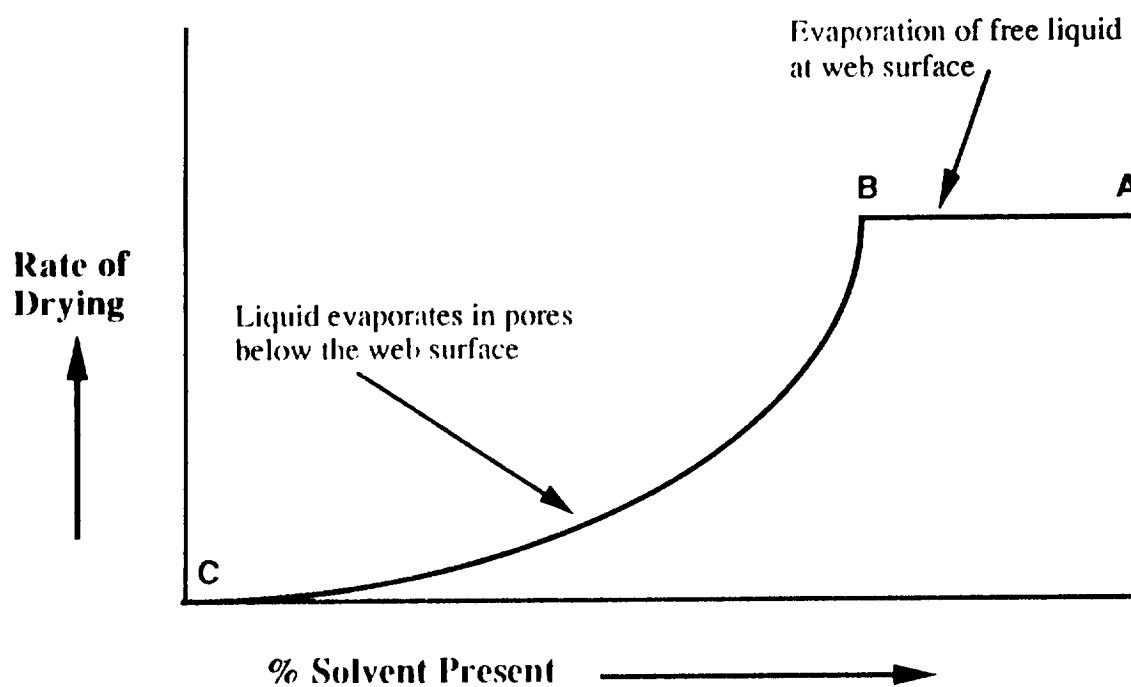


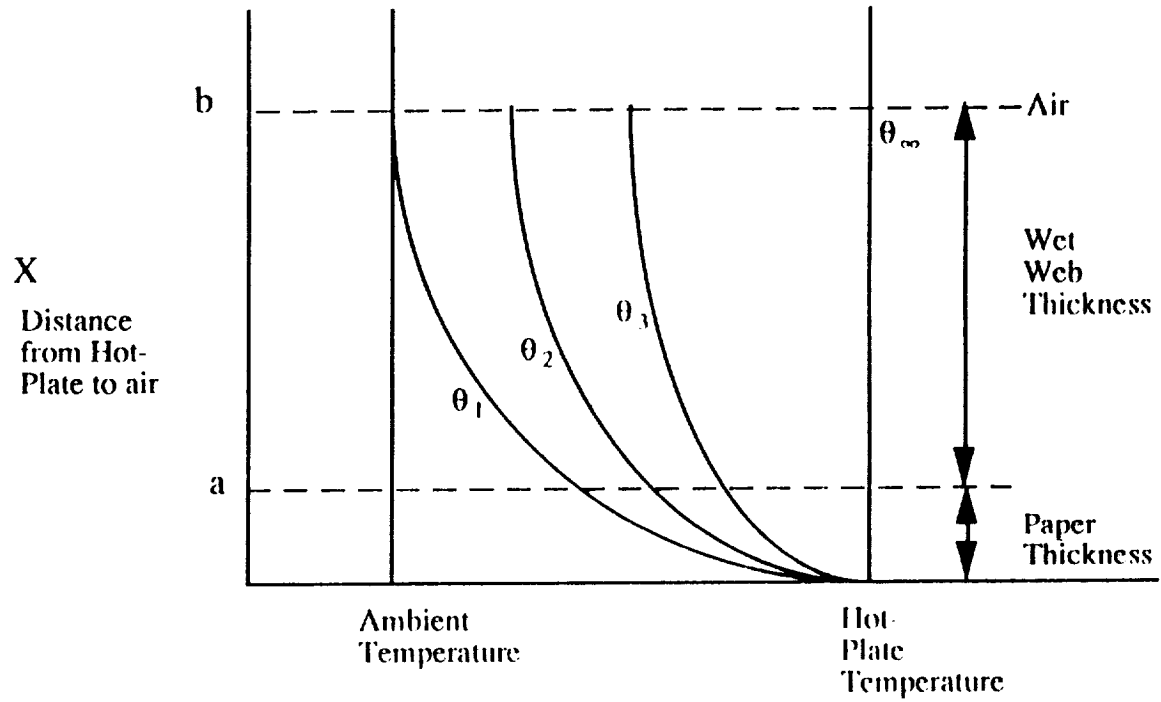
Figure 5. Solution Prepregging Using the Dip Tank Method.

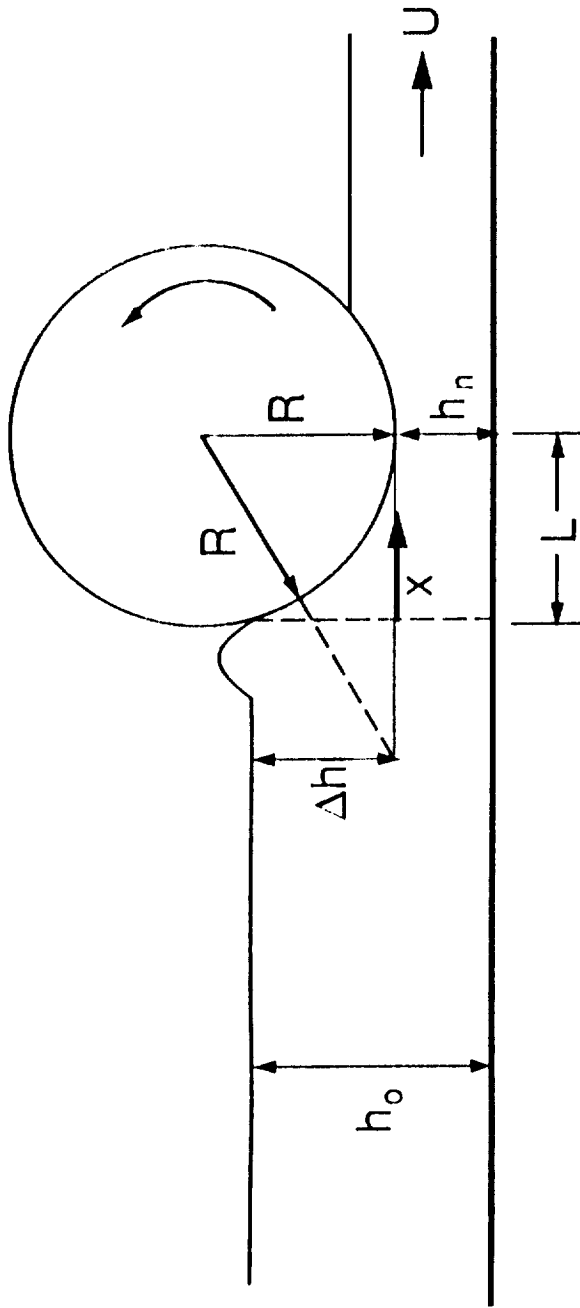


**Figure 6**  
**Rate of Drying As A Function of Solvent Present Within the Web**



**Figure 7**  
**Temperature Variation With  $\lambda$  Through The Thickness Profile**





$$h_o = h_n + L^2 / 2R$$

$$h = h_n + (L - x)^2 / 2R$$

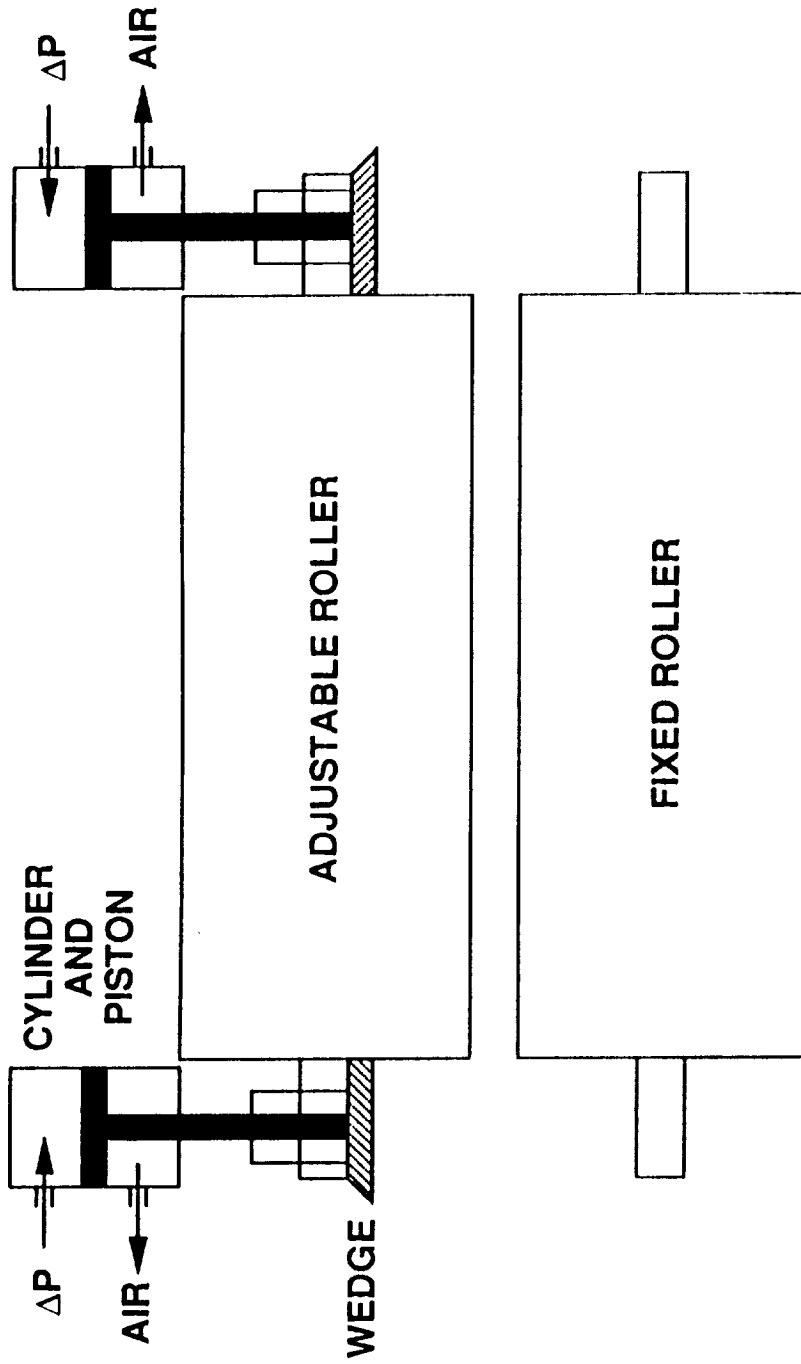
$$P = 4 \mu U R (h - h_n) / g h^2$$

for  $x \geq 0$

where:  $\mu$  is the viscosity

$g$  is the gravitational constant

Figure 8. Schematic Representation of the Calendaring Process.



- **WEDGE INSERTION LIFTS ROLLER CREATING AN OPEN GAP.**
- **WEDGE REMOVAL LOWERS ROLLER ONTO WEB, APPLIED AIR PRESSURE SUBJECTS WEB TO LOAD.**

Figure 9a. Front View of the Nip Roller Operation.

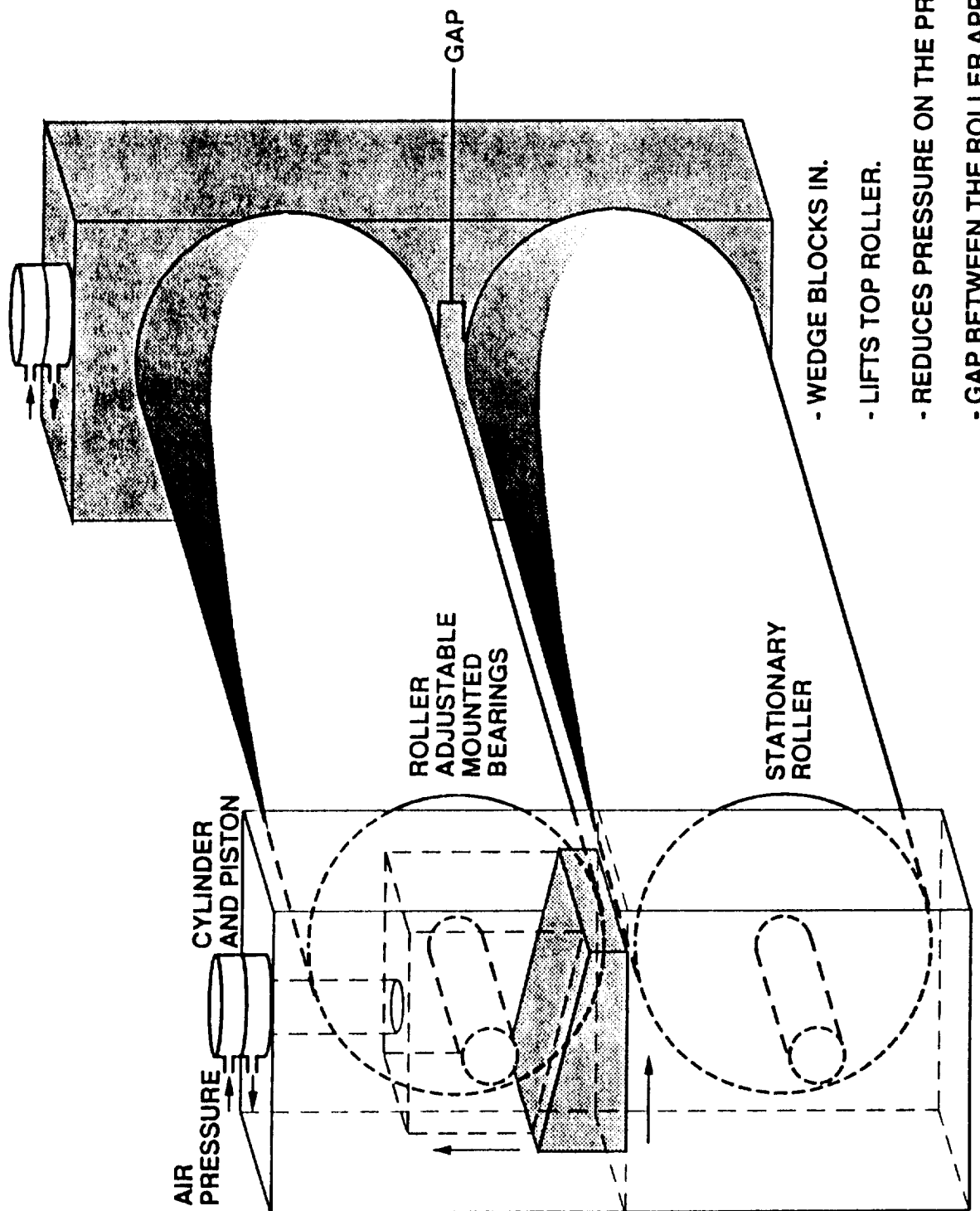
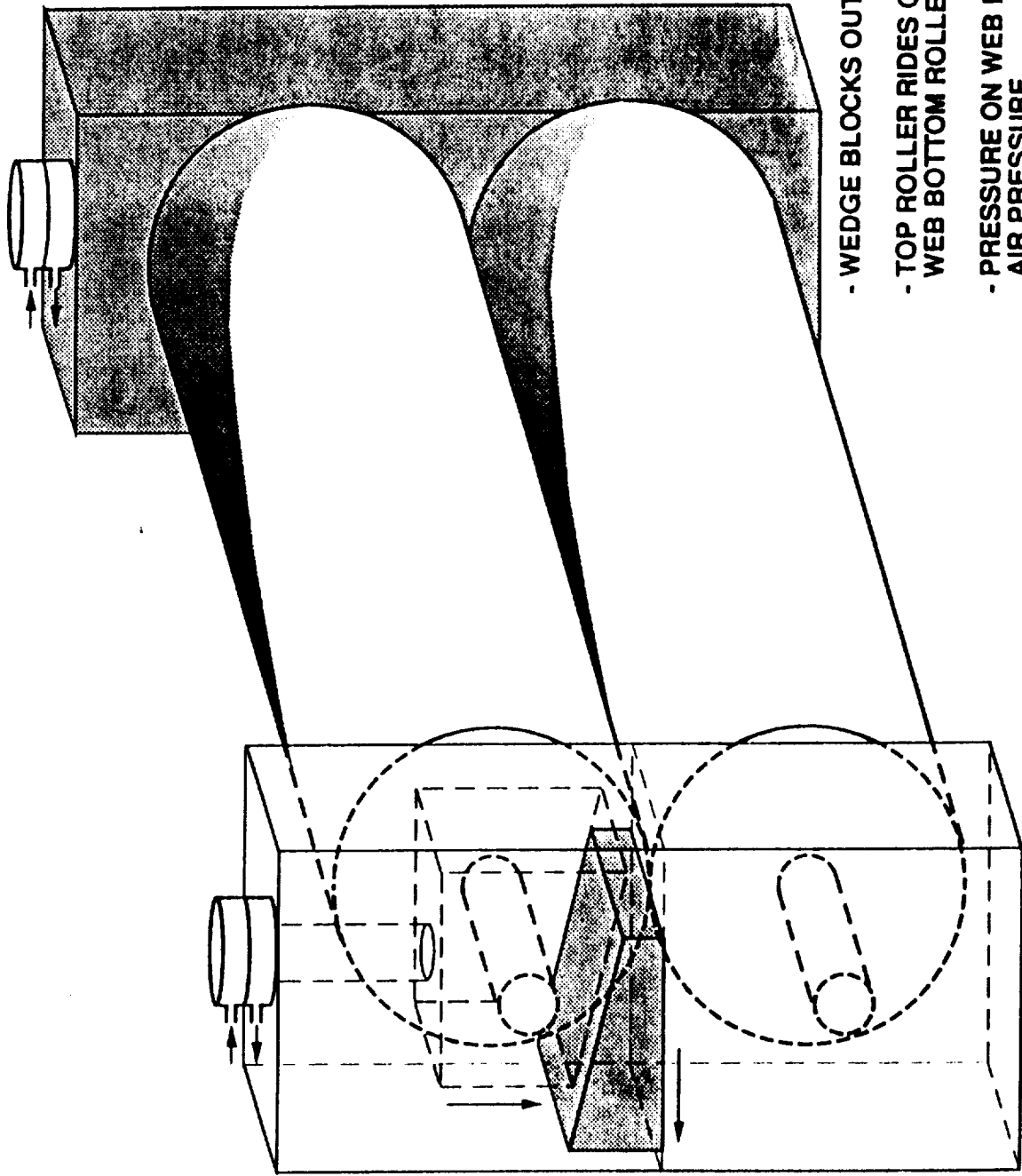


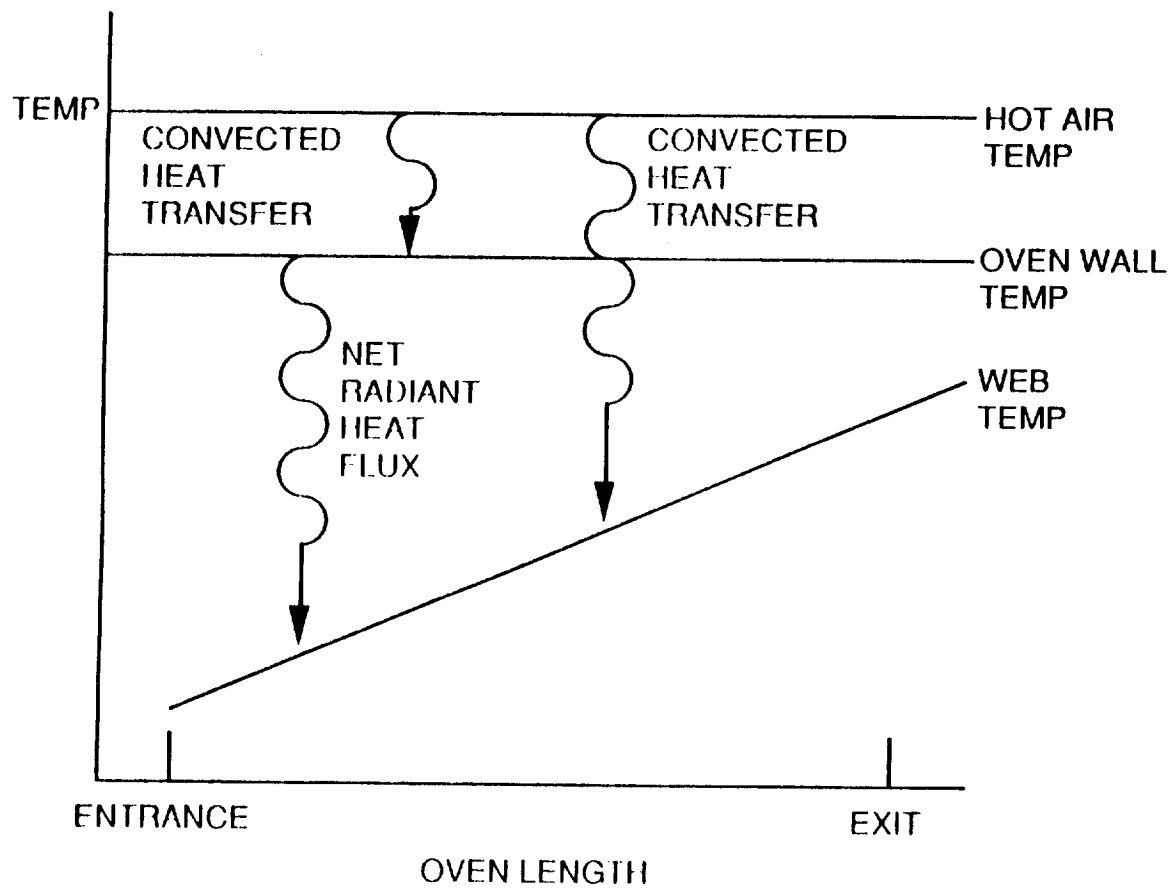
Figure 9b

Three Dimensional View of The Nip Roller Operation With Wedge Blocks Controlling The Gap Size



**Figure 9c**  
**Three Dimensional View of The Nip Roller Operation With Wedge Blocks Out Thereby Closing The Gap**

**Figure 10**  
**Heat Transfer In The Tape Machine Ovens**



# HIGH TEMPERATURE SLED

Side perspective view

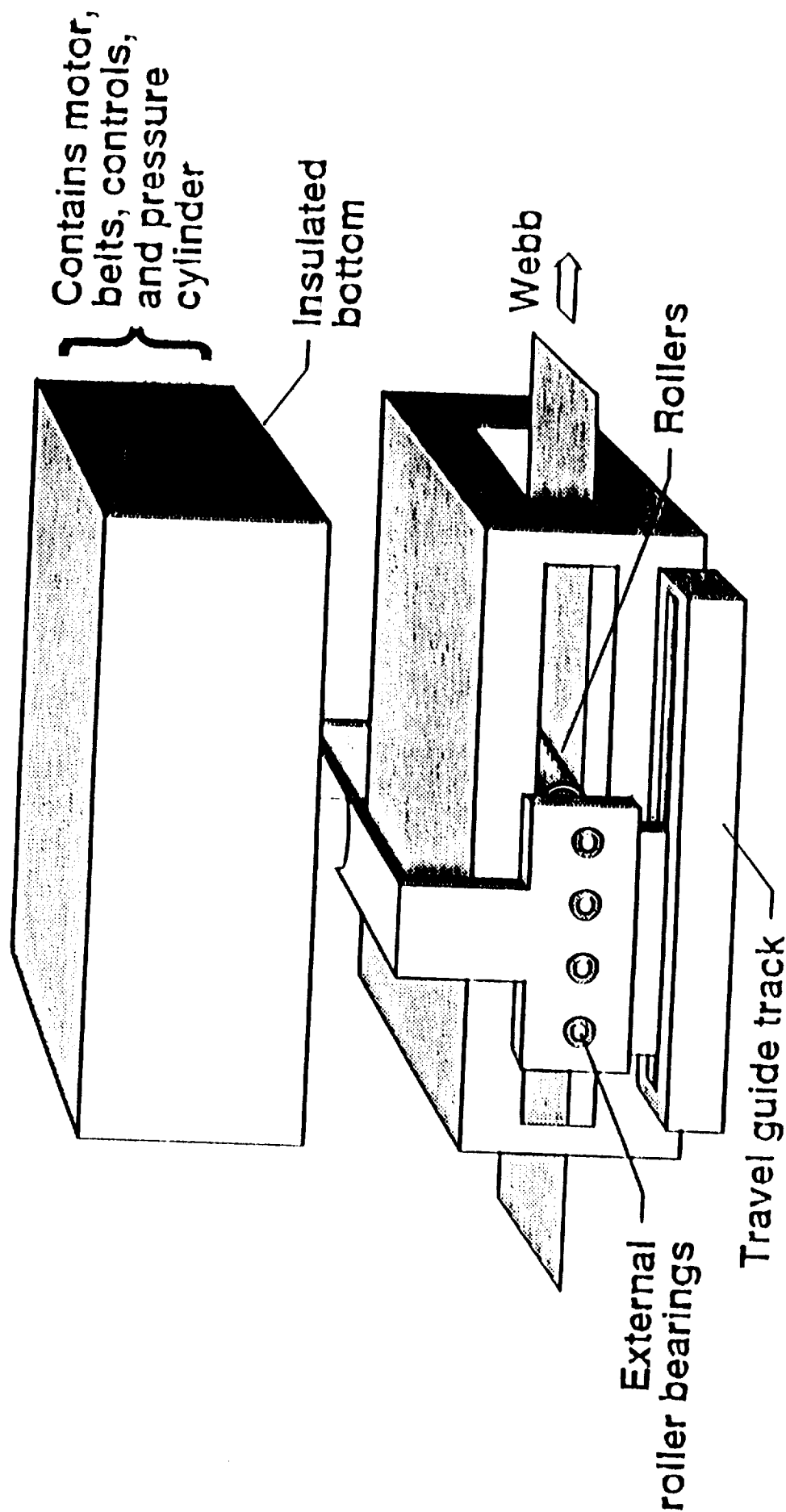
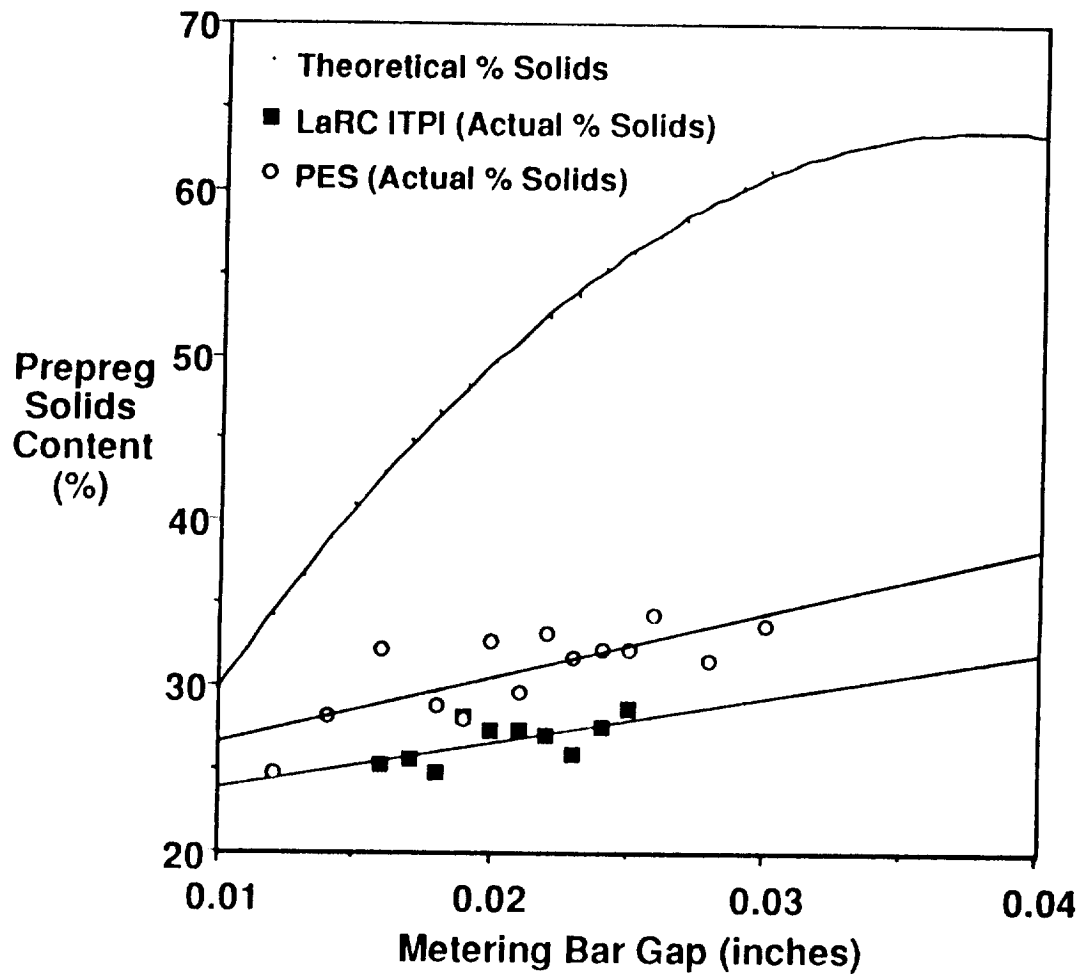


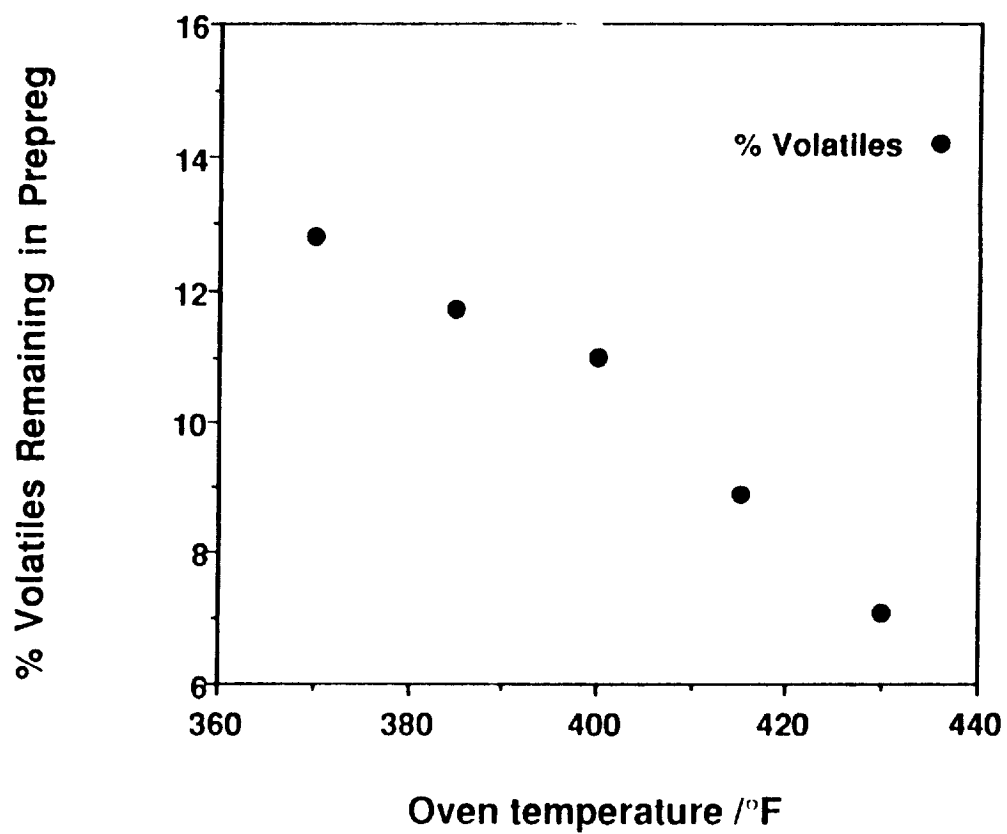
Figure 11  
Schematic Diagram Of The Hot-Sled Attachment-Side View



**Figure 12**  
**Predicted And Actual Percent Solids On LARC ITPI/IM7 Prepreg**



**Figure 13**  
**Removal of Volatiles in LaRC ITPI/IM7 Prepreg**





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13. ABSTRACT (Maximum 200 words) <p>This memorandum presents an introduction to the NASA multi-purpose prepregging unit which is now installed and fully operational at the Langley Research Center in the Polymeric Materials Branch. A description of the various impregnation methods that are available to the prepregger are presented. Machine operating details and protocol are provided for its various modes of operation. These include, where appropriate, the related equations for predicting the desired prepreg specifications. Also, as the prepregger is modular in its construction, each individual section is described and discussed. Safety concerns are an important factor and a chapter has been included that highlights the major safety features.</p> <p>Initial experiences and observations for fiber impregnation are described. These first observations have given great insight into the areas of future work that need to be addressed. Future memorandums will focus on these individual processes and their related problems.</p>				
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